CLIMATE VARIABILITY AND CARBON MANAGEMENT

Margaret S. Torn

510/486-2223
mstorn@lbl.gov

Climate Variability and Carbon Management is a growing interdisciplinary research program in the Earth Sciences Division. The program's main focus is on conducting research to increase the scientific foundation for prediction, impact assessment, and prevention of climate change. In addition, increased understanding of the earth's biogeochemical cycles and climate is needed for many other pressing issues under the purview of DOE and other public agencies, such as stewardship of water resources and the environmental effects of energy use and land use. To that end, we have active projects on climate and hydrology, climate change, a variety of ecological systems and biogeochemical cycles, and carbon sequestration in geologic, oceanic, and terrestrial systems.

One of the strengths of the Climate Variability and Carbon Management Program is its active partnerships with universities, industry, and other research laboratories. A prominent example of such partnerships is our strong link to research activities on the UC Berkeley campus, including the Berkeley Atmospheric Sciences Center.

Last year (2002), within the Program, more than a dozen PIs led projects that included four divisions and more than 62 people at Berkeley Lab. This year (2003), the Program joined with three other divisions (Energy and Environmental Technologies, Engineering, and National Energy Research and Scientific Computing) in developing a proposal for a Lab-wide climate change initiative.

RECENT ACCOMPLISHMENTS

To illustrate a few recent accomplishments by the Climate Variability and Carbon Management Program, below we describe the new regional carbon sequestration alliance we have joined and the advanced research we are doing in the areas of regional climate studies, terrestrial carbon cycling, oceanic carbon cycling, and geologic carbon sequestration.

CARBON SEQUESTRATION PARTNERSHIP

A major new development this year was the establishment of the West Coast Regional Carbon Sequestration Partnership, in which ESD is playing a central role. This is one of seven partnerships recently established by the DOE to evaluate CO_2 capture, transport, and sequestration technologies best suited for different regions of the country. The West Coast Region comprises Arizona, California, Nevada, Oregon, Washington, and the North Slope of Alaska. This partnership constitutes a consortium of over 35 organizations, including state natural resource and environmental protection agencies; national labs and universities; private companies working on CO_2 capture, transportation, and storage technologies (CS&T); utilities; oil and gas companies; nonprofit organizations; and policy/governance coordinating organizations. In an 18-month Phase I project, this partnership will evaluate both terrestrial and geologic sequestration options. There are five major tasks. The first task is to collect data to characterize major CO_2 point sources, the transportation options, and the terrestrial and geologic sinks in the region. These data will be compiled and organized via a geographic information system (GIS) database.

Another task is to address key issues affecting deployment of CS&T technologies, including storage site permitting and monitoring, injection regulations, and health and environmental risks. As part of this, we will conduct public outreach and maintain an open dialogue with stakeholders in carbon CS&T technologies through public meetings, joint research, and education work. At this point, the data and information from the above tasks can be integrated and analyzed to develop supply curves and cost-effective, environmentally acceptable sequestration options, both near- and long-term. Finally, the partnership will identify appropriate terrestrial and geologic demonstration projects consistent with the options defined above, and create action plans for their safe and effective implementation. If suitable demonstration projects can be identified and are approved by DOE, they will be carried out as a Phase II of the Partnership program.

REGIONAL CLIMATE AND WATER RESOURCES

Berkeley Lab's California Water Resources Research Center investigates regional and local hydroclimates for multiple state and federal agencies. A number of regional climate models tend to overestimate precipitation in California, making it difficult to predict water resources and flooding, or to evaluate climate change impacts with confidence. Berkeley Lab analyses showed that these biases arise because models do not represent snow accurately. Simulating the snow pack also has great practical significance, since snow packs store 80% of California's water supply. A climate change analysis for the year 2100 performed by the Lab projects a significant reduction in snow pack for six major watersheds in the state. The related changes in snowmelt and stream flow suggest there will be higher flow, and thus greater flood danger, in the winter, while summer flows, when demand is greatest, will be lower.

TERRESTRIAL BIOSPHERE CARBON CYCLE

Soils contain twice as much carbon as the atmosphere and exchange carbon with the atmosphere at ten times the rate of fossil fuel emissions. We are using advanced isotopic techniques to study the poorly understood rate-controlling processes hidden underground. Berkeley Lab documentation that fine tree roots live five times longer than previously thought is leading to changes in forest ecosystem models and estimates of the amount of carbon pumped belowground by root growth. One of the focal points of carbon cycle research is the vast range of scales—from a single leaf to an entire continent—that must be bridged with measurements and models. Berkley Lab has implemented a coordinated suite of carbon concentration, isotope, and flux measurements in the Southern Great Plains, as part of the DOE Atmospheric Radiation Measurement (ARM) Program. Data streams began flowing to the ARM archives this year. Simultaneously monitoring from crop fields, tall towers, and aircraft, this facility is possibly the best-instrumented site for regional carbon studies in the world.

OCEAN BIOGOECHEMISTRY

Oceans contain more carbon than any other dynamic reservoir on earth, and thus pose a great observational challenge. The pulses of biological productivity are episodic and rapid, and the areas are vast. As a result, we lack understanding of what controls ocean productivity and the export of carbon from the productive photic zone to deeper waters, where it can be stored for long periods. Berkeley Lab has developed the Carbon Explorer, an autonomous float that uses satellite telemetry to report its observations from distant oceans. Last year, Carbon Explorers were deployed as part of the Southern Ocean Iron Experiment to test the effect of iron fertilization. The results suggest that the impact of iron was greater than expected in waters where other essential elements were deficient. These results were made possible by the continuous observations of the Carbon Explorers over an entire year, a data record that would not have been possible with conventional research ships.

GEOLOGIC CARBON SEQUESTRATION

Geologic sequestration of CO_2 below the earth's surface is one of the most promising options for reducing atmospheric CO_2 over the next several decades, because the technologies to capture and store CO_2 are readily available. Berkeley Lab has been exploring a possible new technology—carbon sequestration with enhanced gas recovery, whereby CO_2 is injected into mature natural gas reservoirs to enhance CH_4 recovery while simultaneously storing CO_2 in the reservoir. Simulations suggest that the method is both technically and economically feasible in many cases. Berkeley Lab was also active during the year in leading an international comparison of numerical codes that predict the fate of CO_2 in storage reservoirs, to better understand human health and environmental impacts. We also demonstrated the first use of seismic imaging between two horizontal wells for detailed characterization of a storage reservoir, and the first use of joint electromagnetic and seismic imaging to quantitatively map the saturation of CO_2 in the subsurface for monitoring and verification.

FUNDING AND PARTNERSHIPS

The Climate Variability and Carbon Management Program is funded by a variety of federal and state agencies, and international collaborations. These include the U.S. Department of Energy's Office of Basic Energy Sciences, Office of Fossil Energy, Office of Geological and Environmental Research, and Office of Biological and Environmental Research; the National Aeronautics and Space Administration; the National Science Foundation; the National Oceanographic and Atmospheric Administration; and the Office of Naval Research, as well as the California Energy Commission and CAL-FED.

UNDERSTANDING AND ASSESSING GLOBAL OCEAN CARBON SEQUESTRATION

James K. Bishop

Contact: 510/486-2457, jkbishop@lbl.gov; http://www-ocean.lbl.gov

RESEARCH OBJECTIVES

Over the past century and a half, atmospheric CO_2 concentrations have risen by over 30% from pre-industrial levels. The increase is approximately half the cumulative emission as a result of human activity, with the oceans acting as a major repository for the anthropogenic carbon. This rapid increase in the atmospheric CO_2 has contributed in some measure to the recent warming trends observed worldwide. Understanding the processes that maintain and change the carbon cycle, and developing strategies for managing carbon fluxes and inventories, are national priorities. The following questions are critical: *How does the ocean naturally sequester carbon? How will this change in the future? Could purposeful enhancement of carbon storage in the ocean be an effective way to manage CO_2 in the atmosphere and are such actions safe?*

Biological transformations of carbon in the sea have an important impact on the atmosphere. Marine phytoplankton, whose biomass is renewed entirely every 1 to 2 weeks, consume CO_2 through photosynthesis at a rate of ~50 Pg C yr^{-1} and transport ~10 Pg C yr^{-1} from the surface layer to the deep sea. These fast biological and equally fast physical processes alter the CO_2 distribution in the surface ocean and atmosphere. If we were to disable the "biological carbon pump," then levels of atmospheric CO_2 would rise by 30%. The challenge is to follow such fast processes on a global scale.

Figure 1. Carbon Explorer just prior to deployment in the North Atlantic Ocean by Jim Bishop. UC Berkeley Graduate student Phoebe Lam assisted. The fully robotic float measures temperature, salinity, pressure, particulate organic carbon biomass, light scattering, and carbon sedimentation during its daily transits from kilometer depths to the surface. Data are transmitted to shore in real time for the greater part of one year.

APPROACH

The international project, Argo, is seeding the ocean with thousands of low-cost, long-lived autonomous profiling floats for studying the variability of heat, salinity, and mid-depth circulation of the ocean. Four years ago, we initiated a collaborative effort with Argo scientists to create the first robotic Carbon Explorer—a fully robotic telemetry- and sensor-enhanced version of an Argo float—carrying new optical sensors for characterizing the distribution and fate of marine biology products.

Under Berkeley Lab leadership, Carbon Explorers controlled to cycle between the surface and kilometer depths have been deployed in the subarctic North Pacific (April 2001, February 2003) and in the Southern Ocean surrounding Antarctica (January 2002), where they have remained operational for more than one year in notoriously stormy seas. Three more have just begun observations in the North Atlantic (June 2003).

ACCOMPLISHMENTS

Our North Pacific Carbon Explorers documented the response of marine biota to an iron-deposition event associated with a massive dust storm originating in northeast Asia (Bishop, Davis, and Sherman, 2002).

In the Southern Ocean, Carbon Explorers have quantified an immediate biomass enhancement in response to deliberate iron amendment (Bishop et al., 2002). Also, for the first time, they have documented carbon exported from such experiments into the deep sea. Results have been submitted for publication.

SIGNIFICANCE OF FINDINGS

The development of the Carbon Explorer has truly revolutionized the study of ocean biogeochemistry, by opening an entirely new path for ocean carbon cycle understanding. We have proven an inexpensive method for following biological processes in the ocean, on daily time scales, for the greater part of one year. No limitation prevents implementation of sensors for other carbon components and fluxes on the Carbon Explorer.

The Explorers are inexpensive enough for wide deployment in the oceans to follow the natural carbon cycle. They can also perform observations during small-scale experiments, such as those designed to study ocean ecosystem response to ocean fertilization.

RELATED PUBLICATIONS

Bishop, J.K.B., R.E. Davis, and J.T. Sherman, Robotic observations of dust storm enhancement of carbon biomass in the North Pacific. Science, 298, 817–821, 2002.

Bishop, J.K.B., T.J. Wood, and J.T. Sherman, Carbon Explorer assessment of carbon biomass variability and carbon flux systematics in the upper ocean during SOFeX. EOS Trans Am Geophys Union, 83(47), F799, 2002.

ACKNOWLEDGMENTS

MODELING SUPERCRITICAL CO_2 INJECTION IN BRINE-BEARING FORMATIONS

Christine Doughty, Karsten Pruess, and Sally M. Benson

Contact: Christine Doughty, 510/486-6453, cadoughty@lbl.gov

RESEARCH OBJECTIVES

Geologic sequestration of CO_2 in brine-bearing formations has been proposed as a means of reducing the atmospheric load of greenhouse gases. Numerous brine-bearing formations have been identified as having potential for geologic sequestration of CO_2. One promising setting is the fluvial/deltaic Frio formation in the upper Texas gulf coast, which is the site of an upcoming pilot test of CO_2 sequestration. The objective of this research is to investigate the physical processes controlling the behavior and ultimate fate of CO_2 in the subsurface, to help design the pilot test and to gain a broader understanding of the issues accompanying CO_2 sequestration in brine-bearing formations.

APPROACH

To evaluate CO_2 sequestration scenarios, we use the numerical simulator TOUGH2, which considers all flow and transport processes relevant for a two-phase (liquid-gas), three-component (CO_2, water, dissolved NaCl) system. In the subsurface, supercritical CO_2 forms an immiscible gas-like phase and partially dissolves in the brine.

A three-dimensional numerical model is developed of the pilot test site, a 450 m × 450 m dipping fault block containing several wells that penetrate the 12 m thick brine-saturated sand near the top of the Frio that is our sequestration target. Under the planned sequestration conditions (P = 150 bars, T = 66°C), supercritical CO_2 is strongly buoyant compared to the native brine.

ACCOMPLISHMENTS

We have simulated a number of alternative scenarios for the pilot test, varying three types of model parameters:
- Operational parameters such as injection and monitoring well locations and injection schedule
- Geological features such as the continuity of shale layers, the connectivity of sand channels, and the permeability of faults
- Multiphase flow properties such as relative permeability curves

Simulations show that relative permeability functions have a strong effect on CO_2 plume development. Because most of our knowledge and experience concerning relative permeability for the Frio comes from petroleum reservoirs, in which liquid phases displace a pre-existing gas phase, how to choose appropriate relative permeability functions for supercritical CO_2 injection into a brine-saturated formation is still an open question. Snapshots of the simulated supercritical CO_2 plume (Figure 1) show the impact of relative permeability. For relative permeability functions with large residual gas saturation

S_{gr}, the plume is compact and does not move much under buoyancy forces, because much of the gas is immobile. In contrast, for relative permeability functions with small S_{gr}, the plume is more diffuse. It moves and spreads significantly over time, allowing a much larger fraction of the CO_2 to dissolve in the brine.

SIGNIFICANCE OF FINDINGS

The ability to numerically simulate the complex multiphase flow processes involved in CO_2 injection is critical to developing a good experimental design for the pilot test, just as it will ultimately be for designing successful sequestration operations.
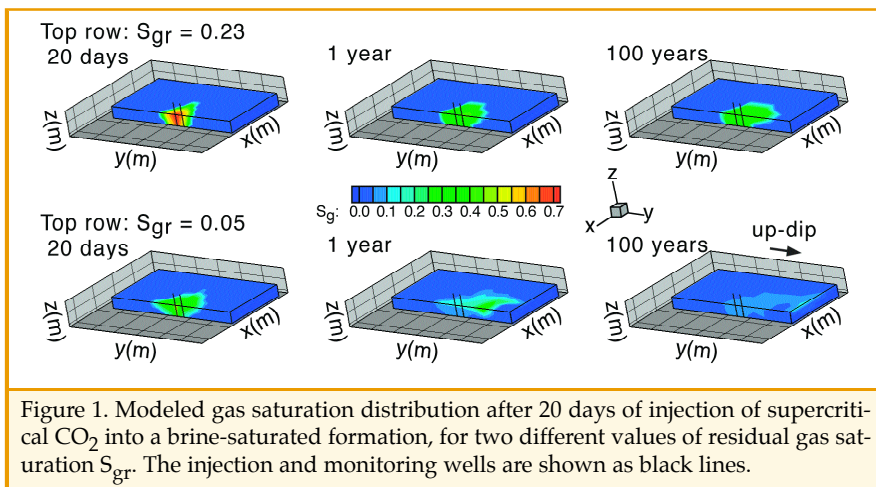

Figure 1. Modeled gas saturation distribution after 20 days of injection of supercritical CO_2 into a brine-saturated formation, for two different values of residual gas saturation S_{gr}. The injection and monitoring wells are shown as black lines.

The residual gas saturation used in the relative permeability functions is a key factor controlling the development of the CO_2 plume. Future laboratory and field work will be directed toward determining appropriate values of S_{gr} for CO_2 injection into brine-bearing formations.

RELATED PUBLICATIONS

Doughty, C., and K. Pruess, Modeling supercritical CO_2 injection in heterogeneous porous media. Presented at TOUGH Symposium 2003, Berkeley Lab., Berkeley, California, May 12–14, 2003.

Related web site: http://www-esd.lbl.gov/GEOSEQ/index.html

ACKNOWLEDGMENTS

TRACKING STABLE ISOTOPES IN A REGIONAL CLIMATE MODEL

Prudence N. Foster, Norman L. Miller, and Don DePaolo
Contact: Norman L. Miller, 510/495-2374, nlmiller@lbl.gov

RESEARCH OBJECTIVES

The ability to track isotopes through the water cycle in a climate model offers the opportunity to test the climate model itself, as well as to learn more about the water cycle. While several global climate models currently have isotope-tracking subroutines, there is to date no regional climate model with isotope tracing. Well-recognized advantages of a regional climate model, over a global one, include the ability to study the natural variability of local water budgets on scales of interest to water planning and ecological impact analyses. Adding tracing routines to the regional model will allow us to study the source of local rainfall and to better understand its sensitivity to climate and land surface changes. Moreover, *isotope* tracking throughout the water cycle will allow an unprecedented ability to test numerical precipitation schemes. Currently, climate models compare their precipitation predictions based primarily on the amount of rainfall. Since rain can develop in a variety of ways, simply getting the amount correct is not a guarantee that the model is actually simulating reality. By comparing the isotopic content of both deuterium and ^{18}O in the rainfall to observations on the scales at which observations can actually be extensively done (i.e., the regional scale), we will have much greater confidence in our rainfall schemes. Such intensive observations are already under way at the DOE Atmospheric Radiation Measurement-Cloud and Radiation Testbed (ARM-CART) site in Kansas (Machavaram et al., 2003), and we plan to make comparisons between our model and these observations.

APPROACH

Our approach has been to add isotope tracers to the community regional climate model MM5 (fifth generation Mesoscale Model). First of all, we have added a set of tracers that exactly copy the water cycle itself. Now, we are working on tracking "colored water"; for example, red water only enters the grid from the water surface of the Gulf of Mexico. We are testing various schemes for tracking the colored water through the processes of surface evaporation, mixing in the planetary boundary layer and cloud physics. The colored water will allow us to understand where water is coming from over the ARM/CART site. Once we have some assurance that these schemes are redistributing the colored water in a manner that we expect, adding the fractionation associated with various phase changes should be trivial. We will then be in a position to validate the model predictions against the observations of precipitation and vapor isotopic values measured in June 2000 at the ARM/CART site.

ACCOMPLISHMENTS

To date, we have completed a map of the water cycle in MM5. We have reproduced the water cycle with an additional vector that can exactly follow the treatment of water or can be manipulated to test code development. We have also prepared the fractionation subroutines that will be needed when the code is ready to address the issue of isotopes.

ACKNOWLEDGMENTS

PERFORMANCE REQUIREMENTS FOR GEOLOGICAL STORAGE OF CO_2

Robert P. Hepple and Sally M. Benson
Contact: Robert P. Hepple, 510/486-5989, rphepple@lbl.gov

RESEARCH OBJECTIVES

The probability that long-term geologic storage or sequestration of CO_2 will become an important climate change mitigation strategy depends on a number of factors, namely (1) public acceptance, (2) the cost of geologic storage compared to other climate change mitigation options, and (3) the availability, capacity, and location of suitable sites. Whether or not a site is suitable will be determined by establishing that it can meet a set of performance requirements for safe and effective geologic storage. Establishing effective requirements must start with an evaluation of how much CO_2 might be stored, and how long the CO_2 must remain underground, to meet goals for controlling atmospheric CO_2 concentrations. These requirements then provide a context for addressing the issue of what is an "acceptable" surface seepage rate.

APPROACH

To address the question, "How much CO_2 might be stored underground and for how long?" we developed zeroth-order estimates for the annual amount of CO_2 that would need to be sequestered to meet atmospheric stabilization targets of 350, 450, 550, 650, and 750 ppmv. We assumed geologic sequestration would be used as a bridging technology, allowing for the gradual phase-out of fossil fuels over a period of up to 300 years. We also assumed that geologic storage constitutes the only mitigation outside of the climate-forcing parameters included in the emissions scenarios (e.g., parameters such as the rates of technology and economic development, and the strength of the movement toward global environmental and sustainability ethics.)

To address a second important question, "What would be an acceptable surface seepage rate?" we first calculated the rate at which CO_2 might seep back to the surface and then compared the calculated seepage to the allowable emissions for atmospheric CO_2 stabilization at each of the five targets. We assumed that the amount of seepage would be proportional to the total amount of CO_2 stored underground at any given time.

ACCOMPLISHMENTS

Figure 1 shows the range of projected storage amounts across the potential stabilization targets, which average between 900 and 2,500 GtC, and it includes estimated storage capacity for comparison. For an annual seepage rate of 0.01% or 10^{-4}/year, the maximum annual seepage never exceeds 0.5 GtC/year for any of the projected sequestration scenarios and would ensure that at least 90% remained effectively sequestered after 1,000 years. For comparison, the total

estimated worldwide volcanic and magmatic degassing is estimated to be 0.07 to 0.13 GtC/year. Because seepage rates less than 0.01% per year meet several criteria for all scenarios, this may be a reasonable long-term global performance requirement for surface seepage.
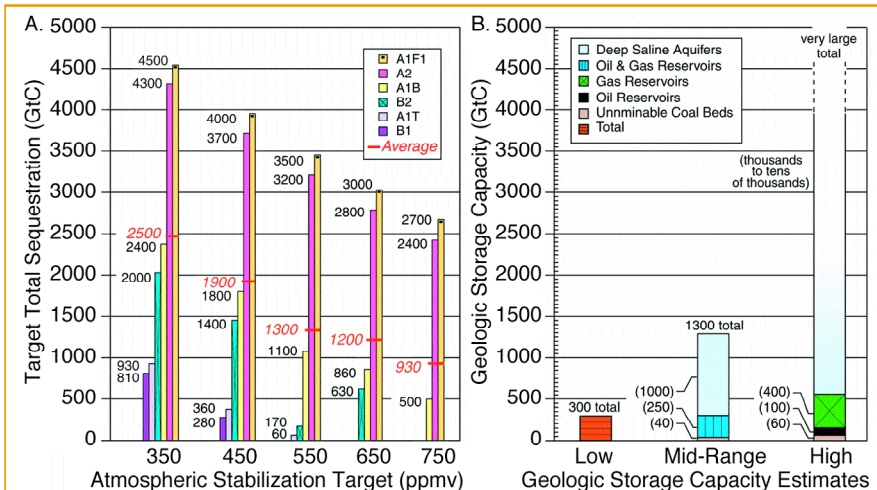

Figure 1. Total target sequestration in Gigatonnes of Carbon (GtC) for each scenario across the range of potential atmospheric stabilization targets in parts per million (ppm) of carbon dioxide.

SIGNIFICANCE OF FINDINGS

According to the results presented here, geologic storage could be an effective method to ease the transition away from a fossil-fuel-based economy over the next several decades to centuries, even if large amounts of CO_2 are stored and some small fraction seeps from storage reservoirs back into the atmosphere.

RELATED PUBLICATIONS

Benson, S.M., R. Hepple, J. Apps, C.-F. Tsang, and M. Lippmann, Lessons learned from natural and industrial analogues for storage of carbon dioxide in deep geological formations. Berkeley Lab Report LBNL-51170, 2002.

Hepple, R.P. and S.M. Benson, Geologic storage of carbon dioxide as a climate change mitigation strategy: performance requirements and the implications of surface seepage. Env. Sci. Tech., 2003 (submitted).

ACKNOWLEDGMENTS

A Mesoscale Analysis of the Impact of Snowpack on Climate Variability in the Sierra Nevada Region

Jiming Jin and Norman L. Miller

Contact: Jiming Jin, 510/486-7551, jimingjin@lbl.gov

RESEARCH OBJECTIVE

Greater than 70% of the annual streamflow in the western United States is derived from snowpack. Hence, accurately forecasting snowpack is essential to this region's economy and well-being, and numerical models are necessary (and power-ful) tools for this purpose. The objectives of this study were to evaluate the snow scheme with an advanced mesoscale model, using observational evidence, and to investigate the impact of snowpack on climate variability in the Sierra Nevada region.

APPROACH

The model used is the fifth-generation Mesoscale Model (MM5) developed by the Pennsylvania State University/National Center for Atmospheric Research. This model was used to generate 12 km resolution results that account for complex topography in the Sierra Nevada. The observed Snow Water Equivalent (SWE) depths for this region were assimilated into MM5 to allow for an investigation into snow evolution and its related processes. This investigation was an approach toward correcting the identified model deficiencies caused in part by the simple snow physics in the land-surface model coupled to MM5. The observed daily SWEs were meas-ured through the automated Snowpack Telemetry system dur-ing the snowmelt season from April 1998 to June 1998.

ACCOMPLISHMENTS

Comparison of observed and simulated SWEs (Figure 1a) indicates that at the 12 km resolution, MM5 poorly represents the snowpack over the Sierra Nevada region during the snowmelt season. At the same time, with the misrepresented snowpack, the model produces a strong warm bias at the near surface (Figure 1b) and exaggerated precipitation (Figure 1c). Subsequently, the observed SWEs (red line in Figure 1a) were incorporated into the model to improve its climate-simulation performance. After the SWE assimilation, the simulated 2 m height air temperature was in very good agreement with observations. In the model, because the assimilated SWE con-sumes a large amount of energy on account of the melting process, the surface skin temperature was reduced, which decreases the upward sensible heat flux. The decreased sensi-ble heat flux supplied less energy to the near surface air and alleviated the warm bias in the 2 m height air temperature. Furthermore, SWE assimilation caused a lowered sensible heat flux as well as a colder surface, leading to weaker outgoing long-wave radiation, reduced air temperature in the lower tro-posphere, and a stabilizing of the atmosphere. The more stable atmosphere restricted atmospheric convections and thus decreased the amplified precipitation.

SIGNIFICANCE OF FINDINGS

This study (Jin and Miller, 2003) indicates that snowpack has a significant effect on near-surface air and precipitation over the Sierra Nevada. Our findings provide a substantial advancement in our understanding of climate variability in the Sierra Nevada region, as well as direction for future model development.
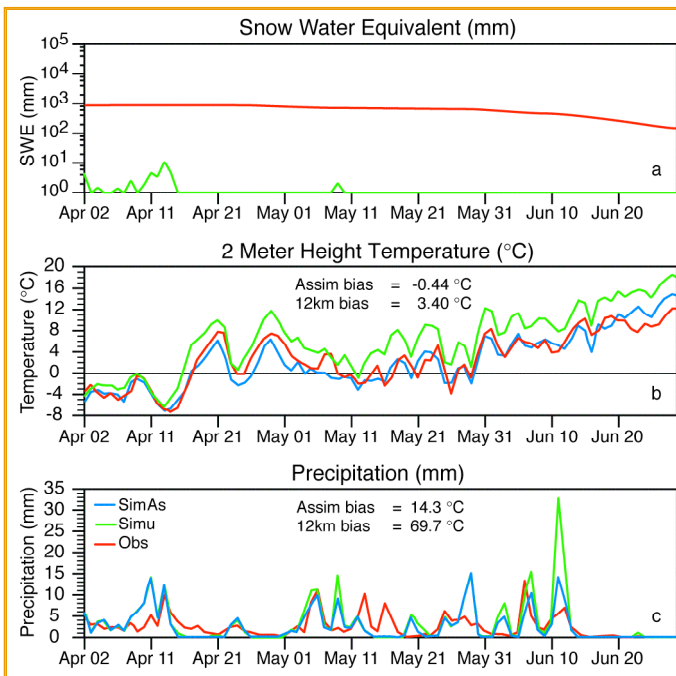

Figure 1. Comparison of simulations and observations aver-aged over the Sierra Nevada region for the period of April 2–June 30, 1998: (a) Snow water equivalent (mm); (b) 2 m height air temperature (ºC); (c) Precipitation (mm). Obs is observa-tions, Simu is the 12 km resolution simulation with no SWE assimilation, and SimuAs is the 12 km resolution simulation with SWE assimilation.

RELATED PUBLICATIONS

Jin, J., and N.L. Miller, A mesoscale analysis of snowpack on cli-mate variability and snowmelt mechanisms in the Sierra Nevada Region. Presented at the PACLIM Conference, April 2003, Pacific Grove, California; J. Hydrometeorology, September, 2003 (submitted).

Jin, J., and N.L. Miller, An analysis of climate variability and snowmelt mechanisms in mountainous regions. J. Hy-drometeorology, September, 2003 (submitted).

ACKNOWLEDGMENTS

IMPACT OF THE ENSO SNOWPACK ON THE WESTERN UNITED STATES: A GLOBAL CLIMATE MODEL STUDY

Jiming Jin and Norman L. Miller
Contact: Jiming Jin, 510/486-7551, jimingjin@lbl.gov

RESEARCH OBJECTIVE

Snowpack is a major water resource in the western U.S.. An accurate forecast of snow amount is essential to water allocation in this region. The objectives of this study are to examine the relationship between El Niño Southern Oscillation (ENSO) and snowpack over this region and to investigate—by comparing observations to simulations from a state-of-the-art global climate model (GCM)—how ENSO affects snow accumulations.

APPROACH

The GCM used in this study is the Community Climate Model Version 3 (CCM3) developed by the National Center for Atmospheric Research (Kiehl et al., 1996). To accurately represent the snowpack in the model, a Snow-Atmosphere-Soil Transfer (Jin et al., 1999) land-surface model with sophisticated snowpack processes was coupled to CCM3. A 45.5-year simulation from December 1949 to May 1995 was generated from this coupled model, which was forced by observed global sea surface temperature (SST) data with year-to-year variations. The observed Snow Water Equivalent (SWE) depths were collected from more than 300 snow-course locations in the western U.S.

ACCOMPLISHMENTS

Figure 1 illustrates the correlation between observed Niño-3.4 SSTs averaged over 120°W–170°W and 5°S–5°N, and SWEs from the model output and observations during winter and early spring for 1950 to 1994. This figure indicates that significant correlations occur in the western U.S. for both simulations and observations, suggesting that ENSO is an important factor causing snow anomalies. Analysis indicates that in the Northwest, the observed anomalous snow patterns are caused by the winter precipitation variability associated with the ENSO, whereas the simulated snow anomalies result from the temperature variations caused by the climate shift. In the Southwest, the simulated positive snowpack anomalies that result from the stronger precipitation are associated with the
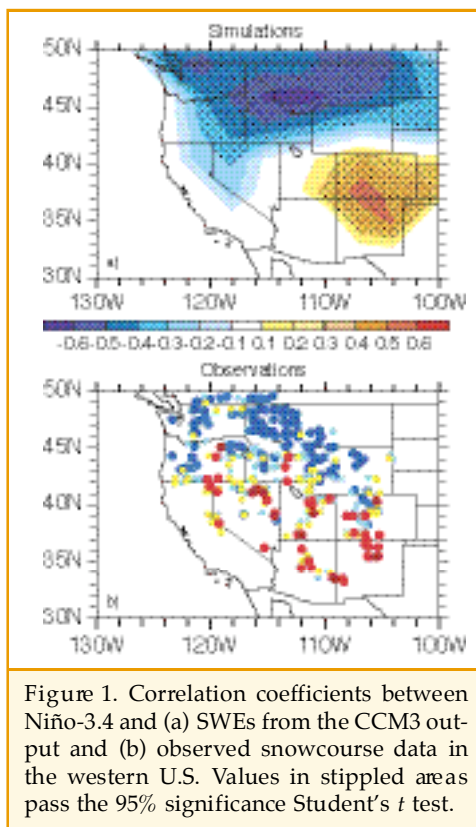

Figure 1. Correlation coefficients between Niño-3.4 and (a) SWEs from the CCM3 output and (b) observed snowcourse data in the western U.S. Values in stippled areas pass the 95% significance Student's *t* test.

warm phase of the ENSO, which is consistent with the observed processes. However, the negative snow anomalies for both simulations and observations appear to have no connections with the tropical Pacific SSTs, which are attributed to the weakened precipitation caused by atmospheric internal variability.

SIGNIFICANCE OF FINDINGS

This study clarifies how the ENSO affects snowpack in the western U.S. and improves our understanding of the mechanism of snow anomalies. The modeled atmosphere in the mid-latitudes incorrectly responds to the tropical Pacific SSTs and shifts the way the air mass gets transported over the Northwest (compared to observations). These findings will greatly benefit climate and water- resources forecasts and future model development.

RELATED PUBLICATIONS

Jin, J., X. Gao, Z. Yang, R.C. Bales, S. Sorooshian, R.E. Dickinson, S. Sun, and G. Wu, One-dimensional snow water and energy balance model for vegetated surfaces. Hydrological Processes, 13, 2467–2482, 1999.

Jin, J., N.L. Miller, S. Sorooshian, and X. Gao, 2003: Impact of ENSO snowpack in the western U.S.: A GCM Study. Presented at the American Meteorological Society Conference, Long Beach, California, Feb. 2003; Journal of Climate, 2003 (manuscript in final preparation).

Kiehl, J.T., J.J. Hack, G.B. Bonan, B.A. Boville, B.P. Briegleb, D.L. Williamson, and P.J. Rasch, Description of the NCAR Community Climate Model (CCM3). NCAR Tech. Note, NCAR/TN-420+STR [Available from the National Center for Atmospheric Research, Boulder, Colorado], 1996.

ACKNOWLEDGMENTS

GEOSTATISTICAL MODELING OF CLIMATE VARIABLES AT REGIONAL SCALES

Phaedon C. Kyriakidis and Norman L. Miller
Contact: Phaedon Kyriakidis, 805/893-2266, phaedon@geog.ucsb.edu

RESEARCH OBJECTIVES

The objective of this research is to develop, and illustrate the application of, a novel geostatistical framework for stochastic modeling of climate variables at regional scales. The developed approach allows for assessing explicitly the uncertainty in hydrological and environmental model predictions due to uncertain climate forcing, with wide applications to risk analysis in related impact assessment studies.

Figure 1. Two alternative simulated realizations of daily precipitation, at 1 km resolution, over the San Francisco Bay Area, for November 13, 1981

APPROACH

The developed geostatistical framework is based on stochastically assimilating (fusing) direct measurements of climate-related variables obtained at monitoring stations, and on ancillary (indirect) information provided by (1) terrain elevation and its derived products (e.g., slope, aspect) and (2) coarse-resolution predictions of climate variables obtained from dynamical downscaling, using a regional climate model. A novel adaptation of stochastic simulation in a space-time context enables the generation of realistic, fine-resolution, alternative synthetic realizations of climate variables at regional scales. These realizations are consistent with (i.e., reproduce exactly) the information available at coarser resolutions available in the form of dynamically downscaled predictions.

ACCOMPLISHMENTS

The development and application of the geostatistical framework for modeling daily precipitation over a region near the San Francisco Bay Area in a space-time context (Case 2 in the Approach section above) is showcased in Kyriakidis et al. (2003). Simulated precipitation realizations at a 1 km resolution were constrained by the available rain-gauge measurements and ancillary terrain-related information. They were shown to reproduce (a) the rain-gauge measurements and their histogram, and (b) a model of their spatiotemporal correlation. Two such simulated realizations are shown in Figure 1. The theoretical proof of mass preservation for the geostatistically derived fine-resolution predictions—that is, the exact reproduction of data available at a coarser resolution (Case 2 above)—is given in Kyriakidis (2003). Current work is focused on a case study illustrating how to apply stochastic simulation of fine-resolution daily precipitation subject to such coarser-resolution data constraints.

SIGNIFICANCE OF FINDINGS

The developed geostatistical framework constitutes a novel approach for the stochastic generation of realistic fine-resolution climate predictions, which can be used in a Monte Carlo setting for risk analysis in environmental and hydrological modeling. When coupled with regional climate model forecasts under future climate change scenarios, the geostatistical framework provides a novel approach to downscaling climate predictions at finer resolutions for more realistic impact assessment studies. The explicit account of the different data supports (i.e., the different volume informed by different types of measurements: rain gauge data versus regional climate model predictions) is a novel modeling characteristic not shared by any of the currently available statistical downscaling methods.

RELATED PUBLICATIONS

Kyriakidis, P.C., N.L. Miller, and J. Kim, A spatial time series framework for modeling daily precipitation at regional scales. Journal of Hydrology, 2003 (in press).

Kyriakidis, P.C., The geostatistical solution of the area-to-point spatial interpolation problem. Geographical Analysis, 2003 (in press).

ACKNOWLEDGMENTS

Isotopic Variations in Atmospheric Moisture in the Great Plains Region

Madhav V. Machavaram, Mark E. Conrad, and Norman L. Miller

Contact: Madhav V. Machavaram, 510/486-5026, mvmachavaram@lbl.gov

RESEARCH OBJECTIVES

The stable isotopic composition of atmospheric water vapor at a given region is determined by its source and subsequent admixture and condensation processes. The most dynamic changes in the water cycle occur in the atmospheric part of the cycle. Thus, the variation in the stable isotope ratios of atmospheric vapor provides critical information about the locally evapo-transpired moisture. In our work, we analyzed atmospheric vapor samples to improve predictability, in part by integrating stable isotope variations into climatological models.

APPROACH

To understand the response of regional hydrology to climatic variations, it is important to quantify the influence of local moisture on precipitation. The deuterium excess (δ-excess)—a measure of the abundance of deuterium (δD) over 18-oxygen (δ^{18}O)—is a valuable tool for estimating the contribution of secondary moisture sources to atmospheric moisture. The *d*-excess in the atmospheric vapors is determined primarily at the oceanic source and altered by admixture of secondary moisture derived through evapo-transpiration. Thus, by measuring the *d*-excess in atmospheric vapors, we can estimate the influence of land-derived moisture.

ACCOMPLISHMENTS (DATA DISCUSSION)

Samples of atmospheric vapor between the earth surface and 3,500 m altitude were cryogenically collected during a flight operation. The stable isotope data for the vapor samples are presented in Figure 1. The samples that were collected below 700 m exhibited much smaller isotopic variation than those from above. The top of the Atmospheric Boundary Layer (ABL) was determined to be at 1,000 m from the surface during the sampling time. The height of the ABL marks the altitude above which the turbulence of the lower troposphere is negligible. Thus, the sample collected at 3,660 m height could

be considered as the atmospheric moisture originating from the Gulf of Mexico.

Assuming that the samples above and below the ABL represent two end members on a linear mixing line, we performed a mixing calculation using the d-excess values, which indicated that approximately 75% of the moisture within the ABL is derived from local sources through evapo-transpiration. Although the contribution of secondary moisture may change diurnally and seasonally, the overall effect of locally derived moisture on precipitation is believed to remain significant over time.

Figure 1. Isotopic variation in the atmospheric vapor samples at various altitudes

SIGNIFICANCE OF FINDINGS

Understanding the influence of locally derived moisture on the water cycle is valuable for improving climatological models. When monitored over longer time scales, such information is extremely useful in documenting the hydrological changes of a region through man-made causes, such as agriculture and urbanization.
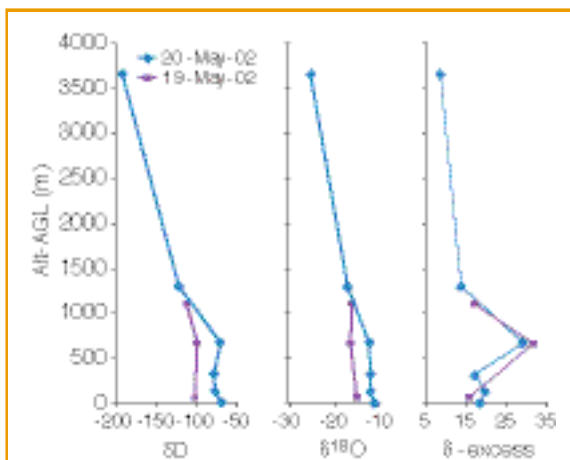
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ACKNOWLEDGMENTS

CROSSWELL AND VERTICAL SEISMIC IMAGING AT THE WEYBURN CO_2 PROJECT

Ernest L. Majer, Dale Cox, Roland Gritto, and Thomas M. Daley

Contact: Ernest L. Majer, 510/486-6709, elmajer@lbl.gov

RESEARCH OBJECTIVE

This work is part of a comprehensive time-lapse seismic monitoring program for monitoring a massive CO_2 flood in a thin fractured carbonate reservoir in EnCana's Weyburn field, located in the Williston Basin, southeast Saskatchewan, Canada. There are two goals to this project: (1) develop and deploy cost-effective technology that could track the detailed changes in CO_2 content as a function of time and (2) aid in the interpretation, validation, and integration of surface seismic data (from 3-C 3-D and 9-C 3-D surface and vertical seismic profiles [VSP]) to obtain an overall understanding of monitoring technologies for CO_2.

APPROACH

An extensive long-term CO_2 miscible injection is being operated by EnCana in its Weyburn field. The first phase of CO_2 injection started in September 2000. The flooding project is expected to expand over the Weyburn field area in the next several years. To determine applicability as well as refine the methods, a comprehensive plan for using geophysical methods for mapping fluid migration and dynamics is being carried out. In addition to the baseline and repeat 3-C 3-D and 9-C 3-D surface seismic and VSP surveys acquired by EnCana and the Colorado School of Mines (CSM) Reservoir Characterization Project, Berkeley Lab is carrying out high-resolution crosswell studies. The higher-resolution borehole data will be integrated with the surface seismic and reservoir engineering models to provide an overall understanding of reservoir definition and the dynamics of fluid migration. The crosswell seismic survey is intended to provide tomographic images of changes in reservoir properties at a meter scale or less. Integrated with the surface seismic survey and VSP, these data will provide proper scaling relationships for understanding overall flow behaviors of the CO_2 fluid at the reservoir dimensions. We will closely study the trade-off between the spatial resolution and spatial coverage of surface methods and borehole methods.

ACCOMPLISHMENTS

The main activities during the last year were a successful implementation of the vertical crosswell and VSP program. We planned
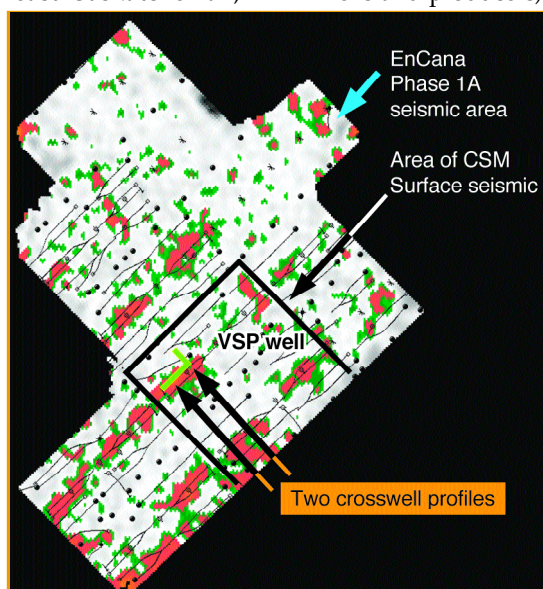

Figure 1. Location of the two vertical crosswell profiles and the VSP acquired in the fall of 2002, relative to the EnCana surface seismic and the CSM surface seismic

and carried out two crosswell seismic profiles in three vertical wells surrounding one of the injection patterns (see Figure 1 for location relative to surface seismic and relative to the injection wells and producers). These profiles provided data parallel and perpendicular to the injector. The objective was to acquire high-resolution crosswell seismic images by using downhole seismic sources and receivers in separate vertical wells, as well as data with sufficient resolution (estimated at a scale of 1 to 2 m) to monitor CO_2 flood front movement and sequestration within the Midale reservoir zones and (most importantly) possible migration from the reservoir into other formations. After the completion of the crosswell, a VSP was run in the center well at the two offsets used for the crosswell. The source was an I/O multi-component vibrator used in the CSM surface seismic work.

SIGNIFICANCE

Both EnCana's and CSM's 4-D results have shown strong seismic anomalies that effectively correlate with the performance in the CO_2 flood front movements and conformance efficiency (supported largely by information such as production data and tracer study). However, EnCana's 4-D surface seismic has also produced some unexpected results. For example, the patterns of the subject VSP and crosswell have so far behaved quite abnormally. It has not yet generated any production response, even after it had received 3.4 billion ft^3 CO_2 (or equivalently 12.3% HCPV [hydrocarbon pore volume], an amount that would have made a normal pattern yield good production response). EnCana's 4-D data further show significant time delay at and below the reservoir layers and seismic energy attenuation, both indicating a significant amount of CO_2 gas accumulated near or above the reservoir. With all the information combined, we believe that a large portion (or even all) of the injected volume may have migrated into the overlying rocks and sequestered there. This likelihood indicates that monitoring this (and other) phenomena using high-resolution crosswell seismic technology may be required to detect zones of uncertainty in CO_2 floods.

ACKNOWLEDGMENTS

UNCERTAINTY ANALYSIS OF CALIFORNIA STREAMFLOW USING MULTIPLE CLIMATE CHANGE SCENARIOS

Norman L. Miller, Kathy E. Bashford, and Eric Strem
Contact: Norman Miller, 510/486-2374, nlmiller@lbl.gov

RESEARCH OBJECTIVES

The objectives of this study are to determine the potential upper and lower bounds of future streamflow response in California, based on a range of climate projections.

APPROACH

Analysis of the range of hydrological response is based on two Global Climate Model (GCM) projections: the Hadley Centre's HadCM2, a warm and wet projection; and the National Center for Atmospheric Research Parallel Climate Model (PCM–B06.06), a cool and dry projection. Three future periods (2010–2039, 2050–2079, 2080–2099) were analyzed. Future watershed mean-area temperature (precipitation) sensitivities were derived from the temperature (precipitation) difference (ratio) between the projected and baseline (1961 to 1990) climatologies. Specified perturbations with increasing temperature (T) and precipitation (P) were also used. Hydrology was simulated using the Sacramento Soil Moisture Accounting Model, for a set of representative basins (Smith, Sacramento, Feather, American, Merced, Kings).

ACCOMPLISHMENTS

Streamflow was analyzed with historical and HadCM2- and PCM-perturbed time series, and specified increments (see Figure 1). During 2010–2039, HadCM2-forced peakflow occurs during the same month with increased peakflow magnitude for the Sacramento (1a), American (1b), and Merced (1c). Peakflow timing during the 2080–2099 period for the American is a month, while the Sacramento timing remains unchanged. The higher elevation Merced has peakflow one month later than the historical, and a secondary peakflow. This secondary high flow results from increased early season snowmelt and a higher snowline, caused by increased temperature.

The relatively cool-dry PCM-forced streamflow significantly decreases during the March-to-July melt season. Peakflow remains close to the historical amount for the Sacramento (2a) and American (2b) for all projected periods, but the Merced (2c) shows an increase during 2010–2039, and then decreases during 2050–2079 and 2080–2099. For these projections, the
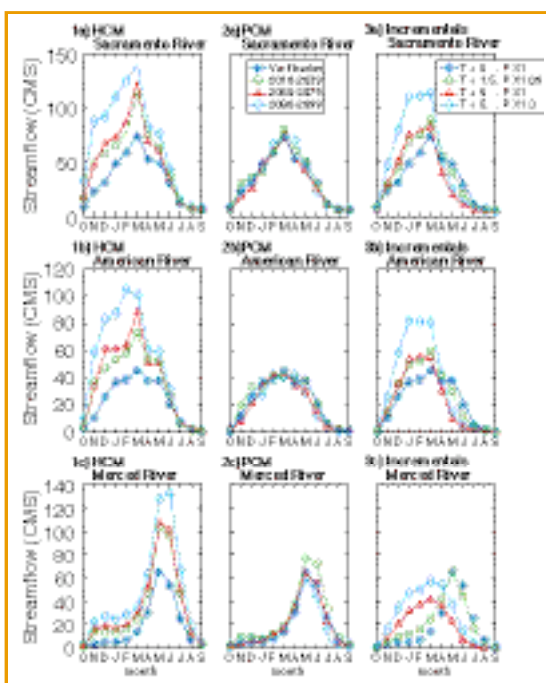

Figure 1. Climatological monthly streamflow at the (a) Sacramento, (b) American, and (c) Merced watersheds for GCM projections: (1) warm-wet and (2) cool-dry. Incremental temperature (0º to 5ºC) and precipitation (0.7 to 1.30 %) are specified for each watershed as well.

American shows a peakflow one month earlier, while the timing of the other two watersheds remains consistent with the historical peakflow timing.

The uniform perturbations bracket projected temperature uncertainties. The 1.5ºC increase and 9% precipitation increase do not change peakflow timing, but increase the October to February peakflow magnitude and slightly decrease the magnitude during the snowmelt period. The peakflow magnitude is higher for the Sacramento (3a) and American (3b), but not for the high-elevation Merced (3c). The Merced peakflow decreases and occurs three months earlier. The extreme scenario represents a high likelihood of more flood events and decreased snowmelt runoff.

SIGNIFICANCE OF FINDINGS

California Sierra Nevada peakflow will likely occur earlier and with increased magnitude. Summer season flow will likely decrease. High-elevation basins are less sensitive to warming, but show a peakflow shift under the incremental changes. The range of outcomes suggests that peakflow magnitudes can shift from 100% increases to 50% decreases.

These results have been applied to water demand and agro-economic analyses (Brekke et al. 2003).

RELATED PUBLICATIONS

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ACKNOWLEDGMENTS

The California Water Resources Research and Applications Center

**Norman L. Miller, K.E. Bashford, G. Brimhall, W.E. Dietrich, J.A. Dracup,
J. Jin, J. Kim, P. C. Kyriakidis, X. Liang, and N.W.T. Quinn**
Contact: Norman Miller, 510/495-2374, nlmiller@lbl.gov

RESEARCH OBJECTIVES

The California Water Resources Research and Applications Center is a NASA-sponsored center designed around a set of integrated activities focusing on California water resources and related impacts. The objectives are to advance our understanding of California hydroclimate variability and change. Core projects include building research partnerships that focus on analysis and educational outreach of hydroclimate impacts on natural systems, society, and infrastructure.

APPROACH

The Center uses dynamical and statistical downscaling schemes within our Regional Climate System Model framework. We produce hydroclimate simulations at short-term, seasonal, and long-term time scales for weather and river flow forecasts, climate change analyses, uncertainty estimates, landslide modeling, water quality monitoring, and climate change assessments of water resources, agriculture, and natural hazards.

Our applications projects include:
- Runoff contaminant monitoring and real-time water-quality monitoring in the San Joaquin Basin
- Contaminant identification and monitoring from Sierra Foothills mine sites
- Development of a dynamic sediment transport and land-slide hazards prediction system
- Snow cover area and water equivalent for California using remotely sensed data and model assimilation
- Geostatistical uncertainty analysis of precipitation and streamflow simulations
- Contributions to impact assessment reports

ACCOMPLISHMENTS

Our Center became a member of the Earth Science Information Partnership, providing value-added climate, weather, streamflow, and impact information to the broad user community, the U.S. National Assessment, and the Intergovernmental Panel on Climate Change reports. We have completed a series of seasonal and multiyear regional climate and streamflow simulations, developed a new statistical downscaling technique for estimating the limits of uncertainty (Kyriakidis et al., 2003), and have used our results as input to the applications listed above. Recent analysis of projected climate and streamflow analysis has been published (Kim et al., 2002) and received national media coverage.

SIGNIFICANCE OF FINDINGS

The climate change and streamflow analyses indicate that the likelihood of extreme weather events will increase, and that night-time temperature will increase at a faster rate than the daytime temperature. An important finding is that warm-wet and cool-dry future climate projections both indicate a 50% decrease in snowpack toward the end of this century. Based on our accomplishments, we received new support from the California Energy Commission, CALFED, and DOE, The California Water Resources Research and Applications Center has become a voice in California climate change assessments, increasing the awareness of potential water resource problems in California and the United States.

RELATED PUBLICATIONS

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Kyriakidis, N.L. Miller, and J. Kim, A spatial time series framework for modeling daily precipitation at regional scales. Journal of Hydrology, 2003 (in press).

ACKNOWLEDGMENTS

THE DOE WATER CYCLE PILOT STUDY: MODELING AND ANALYSIS OF SEASONAL AND EVENT VARIABILITY AT THE WALNUT RIVER WATERSHED

Norman L. Miller, Kathy E. Bashford, Mark S. Conrad, Prudence N. Foster, and Madhav V. Machavaram
Contact: Norman L. Miller, 495-2374, nlmiller@lbl.gov

RESEARCH OBJECTIVES

The DOE Water Cycle Pilot Study is designed to develop the use of water isotopic data (δD, $\delta^{18}O$) to constrain hydroclimate models and test process descriptions and their sensitivity at multiple scales, to better understand water cycle variability. The research objectives are to: (1) evaluate predictions of components of the water budget for several study periods, using a set of nested models with different spatial resolutions, along with archived and new field data from the Walnut River Watershed (WRW); (2) evaluate multiscale water isotope modeling as a means of tracing sources and sinks within and external to the WRW and the Atmospheric and Radiation Measurements Program Southern Great Plains (ARM SGP) site, a representative global climate model grid cell; and (3) identify water-budget-model improvements and data needs over a range of scales.

APPROACH

Water isotopic measurements of precipitation, surface water, soils, plants, and atmospheric water vapor were collected every three months and during the DOE Intensive Observing Period, April 1 to June 30, 2002 (Machavaram et al., this volume). Land-surface modeling compared 1 km fluxes for different modes and for a 50-year simulation. Different wetting and drying conditions caused by different controls were investigated. The Penn State/NCAR Mesoscale Model version 5 (MM5) was advanced with the implementation of water-isotope mass-conservation equations (Foster and Miller, this volume). Multiscale atmospheric simulations using the MM5 and radar-based data have been analyzed and are discussed below.

ACCOMPLISHMENTS

The MM5 6-hour precipitation slightly underestimates for WRW using 4 km resolution during March 1–30, 2000 (Figure 1a). MM5 lags between radar-precipitation onset at March 3. MM5 exhibits considerable accuracy in predicting precipitation occurrences, but shows less accuracy in predicting the precipitation amount. When the 4 km MM5 results are compared to radar precipitation over the entire ARM CART site (Figure 1b), accuracy is improved. MM5 may be underestimating the precipitation in the WRW, but overestimating it in the larger ARM/CART area. This allows for compensating errors, whereas the smaller WRW domain is less forgiving. At 12 km resolution, the MM5 model shows remarkable accuracy in forecasting the total precipitation. At 48 km, the size of the comparison

domain becomes important; the precipitation in the WRW is significantly underestimated, while it is well represented over the entire ARM/CART site. MM5 underestimates the amount of precipitation at 4 km, because it represents the observed variability in precipitation from point-to-point.
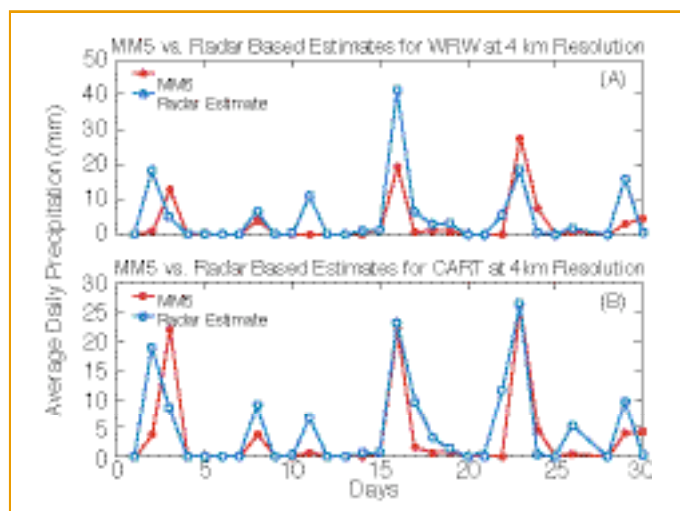

Figure 1. MM5-simulated 4 km precipitation and WSR-88D radar-derived precipitation during March 2000 for (A) WRW domain and (B) the ARM/CART domain.

SIGNIFICANCE OF FINDINGS

During March 2000, there was no convective precipitation, and hence MM5 simulations did not require use of the convective parameterization. The nonconvective precipitation scheme is inadequate at 4 km resolution. It performs better at 12 km, but significantly overestimates variability. This suggests that MM5 may be used during nonconvective situations to predict the amount of precipitation over small watersheds (tens of kilometers) in the U.S. Southern Great Plains during early spring—as long as the resolution is 12 km. Attempts to resolve local-scale precipitation features with MM5 are likely to be biased toward underestimation of precipitation amount.

ACKNOWLEDGMENTS

THE GEO-SEQ PROJECT

Larry R. Myer and Sally M. Benson

Contact: Sally Benson, 510/486-5875, smbenson@lbl.gov

RESEARCH OBJECTIVES

The GEO-SEQ Project is a public-private applied R&D partnership, founded with the goal of developing the technology and information needed to enable safe and cost-effective geologic sequestration of CO_2 by the year 2015. The goals of the project are to:

- Lower the cost of geologic sequestration by (1) developing innovative optimization methods for sequestration technologies, with collateral economic benefits such as enhanced oil recovery (EOR), enhanced gas recovery (EGR), and enhanced coalbed-methane production, and (2) understanding and optimizing trade-offs between CO_2 separation and capture costs, compression and transportation costs, and geologic-sequestration alternatives.

- Lower the risk of geologic sequestration by (1) providing the information needed to select sites for safe and effective sequestration, (2) increasing confidence in the effectiveness and safety of sequestration by identifying and demonstrating cost-effective monitoring technologies, and (3) improving performance-assessment methods to predict and verify that long-term sequestration practices are safe, effective, and do not introduce any unintended environmental impact.

- Decrease the time to implementation by (1) pursuing early opportunities for pilot tests with our private sector partners and (2) gaining public acceptance.

APPROACH

The GEO-SEQ Project consists of four coordinated and interrelated tasks carried out by a multidisciplinary team from eight research organizations: Berkeley Lab, Lawrence Livermore National Laboratory, Oak Ridge National Laboratory, Stanford University, Texas Bureau of Economic Geology, Alberta Research Council, and NITG-TNN, The Netherlands.

The research is conducted with the participation, advice, and cooperation of DOE's National Energy Technology Laboratory and five industry partners: Chevron, Texaco, Pan Canadian Resources, British Petroleum, and Statoil. In addition, through ongoing collaborations and our advisory committee, the team extends to include other universities and a number of public and private research organizations.

ACCOMPLISHMENTS

Highlights of the research conducted to date include:

- Screening criteria for selection of oil reservoirs that could be candidates for co-optimizing CO_2-EOR and CO_2 sequestration were developed.

- Engineering approaches have been developed to increase CO_2 storage, while at the same time enhancing oil recovery.

- Numerical simulations have been carried out to show changes in mineralogy and porosity in a brine-saturated sandstone formation resulting from injection of CO_2 waste streams that are impure (i.e., that contain SO_2, NO_2, and H_2S).

- Numerical reservoir simulations have shown that it is technically feasible to enhance recovery of gas while at the same time sequestering CO_2; the market conditions needed to make the process economical have also been evaluated.

- Software tools have been developed to evaluate the sensitivity of candidate geophysical monitoring methods.

- Effects of hydrocarbons and clay on isotopic compositions need to be taken into account in using isotopic tracers for monitoring reservoir processes.

- State-of-the-art coalbed-methane numerical simulators have been compared, using a set of benchmark problems incorporating increasing levels of complexity.

- An international comparison study of reservoir simulators for oil, gas, and brine formations has been completed.

- Combined crosswell seismic and electromagnetic (EM) surveys were used to quantitatively map gas saturation in a CO_2 EOR pilot in Lost Hills, California.

- The concept of a capacity factor, which could be used to quantitatively compare the sequestration capacity of specific sites, has been developed.

- A pilot brine formation CO_2 injection experiment is being conducted in collaboration with the Texas Bureau of Economic Geology.

SIGNIFICANCE OF FINDINGS

The climate of the earth is affected by changes in radiative forcing caused by several sources, including greenhouse gases (CO_2, CH_4, N_2O). Energy production and the burning of fossil fuels are substantially increasing the atmospheric concentrations of CO_2. One of several proposed strategies to reduce atmospheric emissions is to capture CO_2 from fossil-fuel final power plants and sequester it deep underground. Results from the GEO-SEQ Project are providing methods and information to enable safe and cost-effective geologic sequestration.

RELATED PUBLICATIONS

Publications of the GEO-SEQ Project can be found at http://esd.lbl.gov/GEOSEQ/.

ACKNOWLEDGMENTS

CARBON DIOXIDE FOR ENHANCED GAS RECOVERY AND AS CUSHION GAS

Curtis M. Oldenburg
Contact: 510/486-7419, cmoldenburg@lbl.gov

RESEARCH OBJECTIVES

Natural gas reservoirs are obvious targets for carbon sequestration by direct carbon dioxide (CO_2) injection, because of their proven record of gas production and integrity against gas escape. Carbon sequestration in depleted natural gas reservoirs can be coupled with enhanced gas production by injecting CO_2 into the reservoir as it is being produced, a process called Carbon Sequestration with Enhanced Gas Recovery (CSEGR). In this process, supercritical CO_2 is injected deep in the reservoir while methane (CH_4) is produced at wells some distance away. The active injection of CO_2 causes repressurization and CH_4 displacement to allow the acceleration and enhancement of gas recovery relative to water-drive or depletion-drive reservoir operations. Carbon dioxide undergoes a large change in density as CO_2 gas passes through the critical pressure at temperatures near the critical temperature. This feature makes CO_2 a potentially effective cushion gas for gas storage reservoirs. Thus at the end of the CSEGR process when the reservoir is filled with CO_2, additional benefit of the reservoir may be obtained through its operation as a natural gas storage reservoir. The objective of this research is to demonstrate by numerical simulation the potential sequestration-related uses of CO_2 in natural gas reservoirs.

APPROACH

We have developed a new module called TOUGH2/EOS7C for simulating natural gas reservoirs under CO_2 injection. TOUGH2/EOS7C considers five mass components (water, brine, CO_2, gas tracer, CH_4) and heat. For the gas mixture properties, new real gas mixture subroutines were developed to calculate density and enthalpy departure in the system H_2O-CO_2-CH_4 using the Peng-Robinson equation of state and an accurate gas mixture viscosity model. We use this new module to carry out numerical simulations of CO_2 injection and CH_4 production in model natural gas reservoirs.

ACCOMPLISHMENTS

We have carried out numerous simulations of CO_2 injection, CH_4 production, and natural gas storage with CO_2 as a cushion gas. We present here simulation results for a comparison of native gas (CH_4) and CO_2 cushion gases in a model gas storage reservoir. In Figure 1, we show a schematic of a natural gas storage reservoir showing cushion gas which is not produced, but which compresses upon injection of the working gas (CH_4), and which expands to help produce the working gas (CH_4) upon CH_4 withdrawal. As shown in the pressure vs. time part of the figure, the pressure rise in the reservoir for a given CH_4 injection rate is lower with the CO_2 cushion gas than for a native gas cushion. If the CH_4 injection rate is cut to 70% of the original rate, the pressure rise with a native gas cushion is comparable to the full CH_4 injection rate with CO_2 as cushion gas. In short, more working gas can be injected using a CO_2 cushion than for a native gas cushion. The reason for this is the extreme compressibility of CO_2 around the critical pressure in the 40°C reservoir.

SIGNIFICANCE OF FINDINGS

These simulation results show that CO_2 could be a very effective cushion gas for natural gas storage. Such a use of the reservoir would follow active CO_2 injection that could be used for enhanced gas recovery in a depleting gas reservoir. Our simulations over the last few years show that CO_2 may be a potentially useful gas for both enhancing gas recovery in depleted gas reservoirs, and for use as a cushion gas once the reservoir is filled with CO_2.
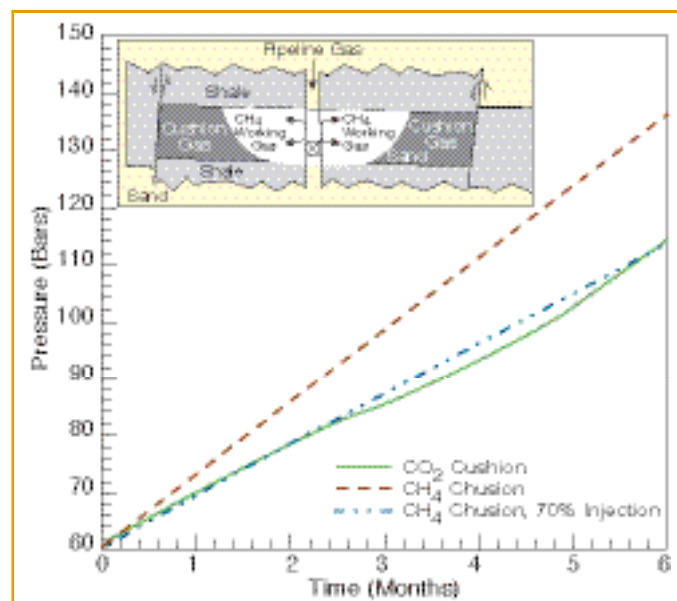

Figure 1. Schematic of natural gas storage and pressure vs. time for one cycle of CH_4 injection with various cushion gases showing the lower pressure rise for CO_2 cushion gas relative to a native CH_4 gas cushion.

RELATED PUBLICATIONS

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ACKNOWLEDGEMENT

COUPLED MODEL FOR CO$_2$ LEAKAGE AND SEEPAGE

Curtis M. Oldenburg and André J.A. Unger
Contact: Curtis M. Oldenburg, 510/486-7419, cmoldenburg@lbl.gov

RESEARCH OBJECTIVES

Geologic carbon sequestration involves the direct injection of large quantities of carbon dioxide (CO$_2$) into deep geologic formations, such as depleted oil and gas reservoirs, saline aquifers, and unminable coal seams. The accumulation of vast quantities of CO$_2$ in the deep subsurface entails the risk that CO$_2$ will leak from the target formation and seep out of the ground. Leakage is CO$_2$ migration away from the primary target formation, whereas seepage is CO$_2$ migration across an interface such as the ground surface. The objective of this research is to investigate the leakage, seepage, and dispersion of CO$_2$.

APPROACH

We have developed a simulation capability called T2CA (TOUGH2 for CO$_2$ and Air) that is an enhancement of the TOUGH2 reservoir simulator. T2CA includes both subsurface and atmospheric surface layer regions. The approach for the subsurface follows the standard multicomponent and multiphase methods in TOUGH2. For the surface layer, we assume a logarithmic velocity profile to represent time-averaged winds. For atmospheric dispersion, we use the Pasquill-Gifford dispersion curves and Smagorinski Model to estimate dispersivities. The logarithmic velocity profile is specified by setting suitable permeabilities and pressure boundary conditions and setting porosity equal to one. In this way, T2CA can handle coupled subsurface–surface-layer flow and transport. This approach is valid for the case in which CO$_2$ concentrations in the surface layer are low and mixing is passive. This appears to be a reasonable assumption for the leakage rates expected in geologic sequestration.

ACCOMPLISHMENTS

We have simulated cases of subsurface, surface layer, and coupled flow and transport using T2CA. For the subsurface, we have carried out a sensitivity analysis where various properties of a thick unsaturated zone were varied. In the surface layer, we have carried out verification studies in which we compared our approach with a commercial fluid-dynamics code and observed good agreement. In Figure 1, we present coupled flow results for wind speed equal to 1 m s^{-1} at a height of 2 m.

SIGNIFICANCE OF FINDINGS

As shown in Figure 1, CO$_2$ concentrations can reach high levels in the subsurface, but are quickly diluted in the surface layer. This result has significance in two important areas of carbon sequestration research: (1) CO$_2$ sequestration monitoring and verification; and (2) health, safety, and environmental (HSE) risk assessment. For monitoring and verification, our results show that detection is more likely if instruments are placed in the subsurface or in trenches. For HSE risk assessment, our results point to the potentially significant risks of subsurface enclosed spaces, such as basements.
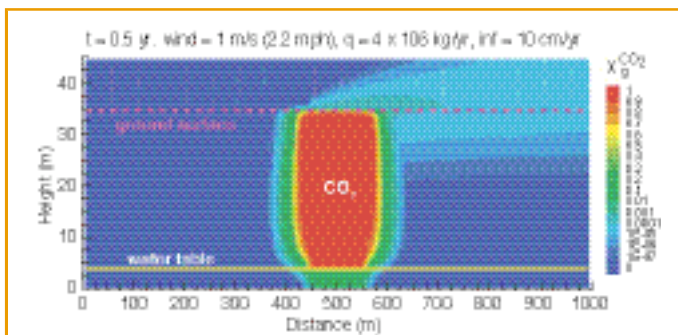
RELATED PUBLICATION

ACKNOWLEDGMENT

Figure 1. CO$_2$ mass fraction and gas velocity vectors for a CO$_2$ leakage scenario involving a source at the water table, with flux equal to 1×10^{-5} kg m^{-2} s^{-1} and wind of 1 m s^{-1} at a height of 2 m from the ground

NONISOTHERMAL EFFECTS DURING CO_2 LEAKAGE FROM A GEOLOGIC DISPOSAL RESERVOIR

Karsten Pruess
Contact: 510/486-6732, k_pruess@lbl.gov

RESEARCH OBJECTIVES

There is general consensus in the scientific community that geologic disposal of CO_2 into saline aquifers would be made at supercritical pressures, P > Pcrit = 73.82 bars. However, CO_2 escaping from a storage reservoir may migrate upwards towards regions with lower temperatures and pressures, where CO_2 would be in subcritical conditions. An assessment of the fate of leaking CO_2 requires the capability to model not only supercritical but also subcritical CO_2, as well as phase changes between liquid and gaseous CO_2 in subcritical conditions.

APPROACH

A new fluid property module was written for the general-purpose TOUGH2 simulator to represent mixtures of brine and CO_2 in all possible phase states, including gaseous, liquid, and supercritical CO_2, as well as an aqueous phase and solid precipitate. This was applied to a study of leakage behavior in a simplified hypothetical model system.

ACCOMPLISHMENTS

Starting from a typical geothermal gradient of 30°C/km in continental crust and hydrostatic pressures, the response to leaking CO_2 entering a vertical channel with elevated permeability at 1,000 m depth was simulated in 2-D cylindrical geometry. A first simulation case was run with an average land surface temperature of 5°C. In this case, the initial T,P-profile intersected the CO_2 saturation line, suggesting that liquid CO_2 will boil as it rises.

The system behavior observed in the simulation can be summarized as follows. Some of the CO_2 dissolved in water, but most of it formed a separate liquid phase. The liquid rose and started vaporizing at about a 630 m depth. Vaporization was partial and gave rise to evolution of a three-phase zone: aqueous—liquid CO_2—gaseous CO_2. Latent heat transfer during phase change caused considerable cooling of the rock, which allowed the liquid front to advance upward (see Figure 1). Over time, the

Figure 1. Advancement of liquid front, and frontal temperature, as a function of time

three-phase zone became several hundred meters thick. Upflow across the three-phase zone was impeded by interference between the phases. As a consequence, CO_2 discharge at the land surface was more dispersed than it otherwise would have been.

The simulation stopped after 391.2 years as freezing conditions were approached. This stoppage occurred because the fluid-property treatment adopted here has no provisions to deal with phase change from liquid water to ice or solid hydrate phases.

Another simulation was run for a larger land-surface temperature of 15°C, in which the initial T,P-profile did not intersect the CO_2 saturation line. Although the rising CO_2 was not subject to phase change, there were nevertheless strong cooling effects because specific enthalpy of CO_2 increases upon depressurization. Over time, temperatures declined and a three-phase zone evolved, similar to what was observed for lower land-surface temperature.

SIGNIFICANCE OF FINDINGS

Upward migration of CO_2 from a geologic disposal reservoir is accompanied by strong heat-transfer effects and gives rise to the evolution of a thick and broad three-phase zone.
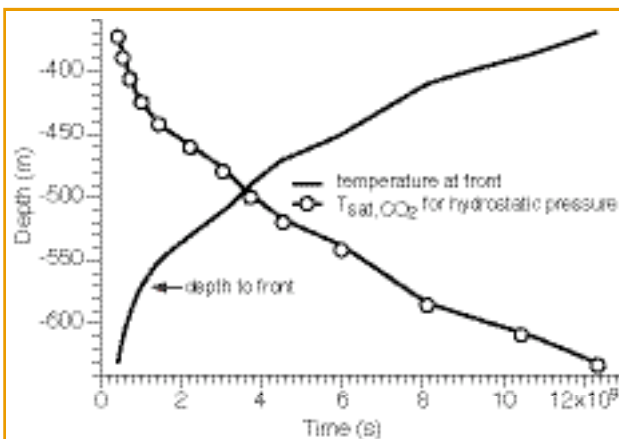
RELATED PUBLICATION

Pruess, K., Numerical simulation of CO_2 leakage from a geologic disposal reservoir, including transitions from super- to subcritical conditions, and boiling of liquid CO_2. SPE Journal, 2003 (submitted). Berkeley Lab Report LBNL-52423.

ACKNOWLEDGMENTS

EFFECT OF WHEAT HARVEST ON REGIONAL CLMATE: EXPERIMENTS WITH THE COUPLED MM5-ISOLSM MODEL

William Riley, Heather S. Cooley[1], and Margaret S. Torn
[1]University of California, Berkeley
Contact: William Riley, 510/486-5036, wjriley@lbl.gov

RESEARCH OBJECTIVES

Surface energy and water fluxes are tightly coupled with CO_2 exchanges between the ecosystem and atmosphere. Other surface-to-atmosphere trace-gas exchanges of interest in climate change research are also strongly impacted by surface energy exchanges. Further, land-use change has considerable impact on the surface ener-gy balance and therefore the exchanges of these trace gases. To investigate these issues at the regional scale, we have cou-pled the meteorological model MM5 with ISOLSM (Riley et al., 2002; 2003), a land-surface model that, in addition to simulating the surface energy balance, predicts gaseous and aqueous fluxes of climatically relevant species within the soil and between the ecosystem and atmosphere.

APPROACH

Here we describe a modeling inves-tigation of the impact of the winter wheat harvest on surface fluxes, air tem-peratures, and regional climate in the Southern Great Plains Atmospheric Radiation Measurement–Cloud and Radiation Testbed (ARM–CART) region (Cooley et al., 2003). We chose to study the winter wheat harvest because wheat accounts for about 20% of the ARM-CART land area, and the fields are typi-cally harvested within a two-week peri-od. Two harvest dates, bracketing the interannual range in the region, were simulated and compared: "early harvest" on June 4 and "late harvest" on July 5.

ACCOMPLISHMENTS

Differences in latent and sensible heat fluxes, 2 m air and soil temperatures, and precipitation varied over the two-month sim-ulation. During the first three days after harvest, midday latent heat fluxes increased substantially, and much of the near-surface soil moisture was lost via evaporation. Over about the next two weeks, midday latent heat fluxes decreased, as transpiration was eliminated and evaporation was sharply reduced in the harvested area. The concurrent increase in sensible heat fluxes resulted in midday air-temperature increases of about 1°C. After the second harvest, air temperature and soil

moisture and temperature levels rapidly converged between the two scenarios, indicating that the system has relatively little memory of the early harvest.

Changes within the harvested areas are substantially larger than the regionally averaged results (Figure 1); further, there are some edge effects. For example, latent heat fluxes increased and sensible heat fluxes decreased in areas adjacent to the harvest because of the drier air being advected from the harvested region.

SIGNIFICANCE OF FINDINGS

Simulations of the winter wheat har-vest in the ARM-CART region indicate that coherent harvesting substantially impacts regionally averaged and local surface conditions and climate. These findings highlight the importance of land use change on climate; in the near future we will use the model to study interac-tions between land use changes and the changing global climate.

RELATED PUBLICATIONS

Cooley, H.S., W.J. Riley, and M.S. Torn, Effect of harvest on regional climate and soil moisture and temperature, in Chapman Conference on Ecosystem Interactions with Land Use Change, Santa Fe, NM, 2003.

Riley, W.J., C.J. Still, M.S. Torn, and J.A. Berry, A mechanistic model of $H_2^{18}O$ and $C^{18}OO$ fluxes between ecosystems and the atmosphere: Model description and sensitivity analy-ses, Global Biogeochemical Cycles, 16, 1095-1109, 2002.

Riley, W.J., C.S. Still, B.R. Helliker, M. Ribas-Carbo, S. Verma, and J.A. Berry, Measured and modeled $\delta^{18}O$ in CO_2 and H_2O above a tallgrass prairie, Global Change Biology, 2003 (in press).
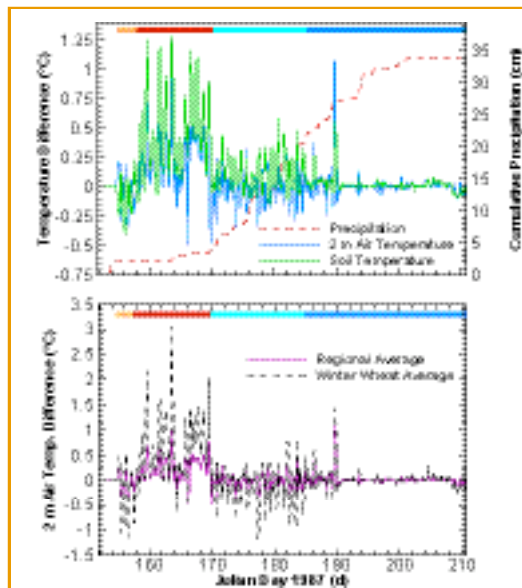
ACKNOWLEDGMENTS

Figure 1. (a) Differences (relative to the late har-vest scenario) in cumulative precipitation and in 2 m air and soil temperatures averaged over the ARM-CART region show four distinct time peri-ods (solid bars at top). (b) Differences in 2 m air temperature in the harvested area and averaged over the entire region. Air temperatures were substantially higher in the harvested area.

Using ^{18}O to Partition Ecosystem Carbon Exchanges: Impact of the Near-Surface δ^{18}O Value of Soil Water on the δ^{18}O Value of the Soil-Surface CO_2 Flux

William J. Riley
Contact: 510/486-5036, wjriley@lbl.gov

RESEARCH OBJECTIVES

The ^{18}O content of atmospheric CO_2 has been proposed as a means to partition site-level measured net ecosystem carbon fluxes into component gross fluxes and, at the global scale, to estimate the regional distribution of CO_2 fluxes. However, these approaches require accurate prediction of the δ^{18}O value of the soil-surface CO_2 flux (δF_s). This work aims to better characterize and to improve the computational efficiency of models used in global and regional simulations.
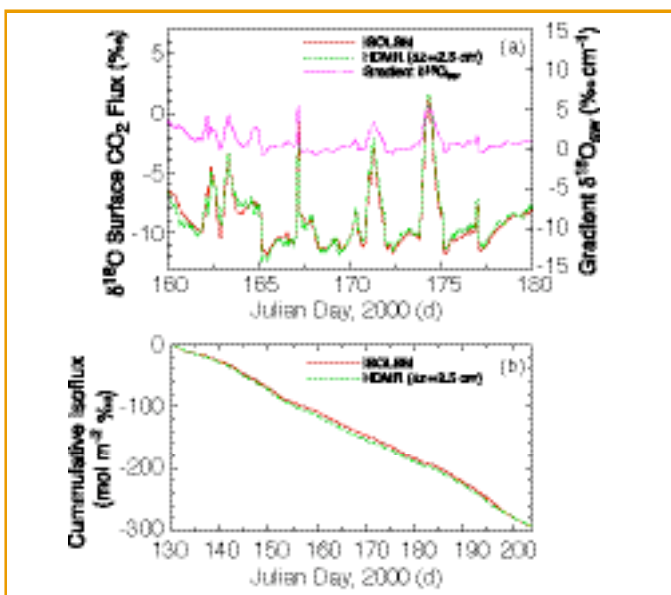
Figure 1 (a) δF_s as predicted by ISOLM and the HDMR approach. Also shown is the gradient in δ_{sw} over the top 15 cm. (b) Cumulative isoflux from the soil to the atmosphere for the two approaches. The error the three-month growing season was 0.2%.

APPROACH

The net $C^{18}OO$ surface flux depends, nonlinearly, on the depth-dependent δ^{18}O value of soil water (δ_{sw}), soil moisture and temperature, soil CO_2 production, and the δ^{18}O value of above-surface CO_2 (Riley, 2003a). We developed ISOLSM (Riley et al., 2002; Riley et al., 2003b) to simulate these processes within an established land surface model (LSM1). ISOLSM simulates the ^{18}O content of canopy water vapor, leaf water, and vertically resolved soil water; leaf photosynthetic $C^{18}OO$ fluxes; CO_2 oxygen isotope exchanges with soil and leaf water; soil CO_2 and $C^{18}OO$ diffusive fluxes (including abiotic soil exchange); and ecosystem exchange of $H_2^{18}O$ and $C^{18}OO$ with the atmosphere. Since ISOLSM is a computationally expensive model, we applied a high-dimension model representation (HDMR) technique to efficiently predict δF_s.

ACCOMPLISHMENTS

Our results indicate that the HDMR approach is very accurate and about 100 times faster than the full numerical solution of the $C^{18}OO$ surface flux, making it appropriate for regional and global simulations. We successfully tested the HDMR approach over a growing season at a C_4-dominated tallgrass prairie site, and then used the model to investigate the factors important in determining δF_s. The top panel of the figure shows comparisons between the full numerical model and the HDMR approach for a 20-day period. The bottom panel shows comparisons of the cumulative isoflux from the soil surface over the full season; the error over the growing season was less than 0.2%. The largest changes in δF_s occur when gradients in the top 5 cm are large. These conditions typically occur when soil evaporation is large, i.e., following precipitation.

SIGNIFICANCE OF FINDINGS

Simulation results indicate that δF_s is dependent on the δ^{18}O value of soil water in the top few centimeters of soil. These results indicate that recent approaches to estimating global distributions of the surface $C^{18}OO$ flux are problematic and demonstrate the importance of accurately resolving near-surface δ_{sw}. Also, the development of the HDMR approach allows for accurate and computationally affordable simulations of regional and global distributions.

RELATED PUBLICATIONS

Riley, W.J., Impact of the near-surface δ^{18}O value of soil water on the δ^{18}O value of the soil-suface CO_2 flux: Application of a high-dimension model representation technique. GRL, 2003a (submitted).

Riley, W.J., C.J. Still, M.S. Torn, and J.A. Berry, A mechanistic model of $H_2^{18}O$ and $C^{18}OO$ fluxes between ecosystems and the atmosphere: Model description and sensitivity analyses. Global Biogeochemical Cycles, 16, 1095–1109, 2002.

Riley, W.J., C.S. Still, B.R. Helliker, M. Ribas-Carbo, S. Verma, and J.A. Berry, Measured and modeled δ^{18}O in CO_2 and H_2O above a tallgrass prairie. Global Change Biology, 2003b (in press).

ACKNOWLEDGMENTS

A Noniterative Model to Compute CO₂-H₂O Mutual Solubilities at 12–100°C and up to 600 Bar

Nicolas Spycher and Karsten Pruess

Contact: Nicolas Spycher, 510/495-2388, nspycher@lbl.gov

RESEARCH OBJECTIVES

Evaluating the feasibility of CO_2 geologic sequestration requires intensive numerical simulations of multiphase fluid flow. These simulations require the calculation of pressure-temperature-composition (P-T-X) data for mixtures of CO_2 and H_2O under moderate pressures and temperatures at which a CO_2-rich phase (gas or liquid) and an H_2O-rich liquid coexist. The objective of this study is to develop correlations to calculate efficiently and accurately the composition of these phases at equilibrium, for implementation into numerical simulations of water-CO_2 flows.

APPROACH

We compiled a large number of published experimental data on the mutual solubility of CO_2 and H_2O over our P-T range of interest into a database (403 data points). We then developed a solubility model, based on equating chemical potentials, and used a modified Redlich-Kwong (RK) equation of state to calculate fugacity coefficients for CO_2 and H_2O in the compressed gas phase. The mixing rules implemented in this model have a standard form but assume infinite H_2O dilution in the CO_2-rich phase. This allowed the use of a noniterative algorithm for computing mutual solubilities at given pressures and temperatures. RK parameters for pure CO_2 were obtained by fitting the equation of state to reference P-T data. Inverse modeling (PEST-ASP v5.0) was then used to calibrate the solubility model to the entire database of experimental P-T-X data, yielding RK parameters for the mixture, as well as aqueous solubility constants for gas and liquid CO_2 as functions of temperature and pressure. Water fugacities needed in the model were taken directly from literature sources.

ACCOMPLISHMENTS

A new model was developed to compute the mutual solubilities of CO_2 and H_2O at 12–100°C and up to 600 bar. The solubility model uses an efficient noniterative algorithm and reproduces experimental data typically within a few percent (Figure 1). The solubility model can be easily extended to moderately saline solutions, using existing activity coefficient models.

SIGNIFICANCE OF FINDINGS

Other CO_2-H_2O solubility models have been published in the literature. However, these models either do not cover our entire P-T range of interest or involve complex correlations
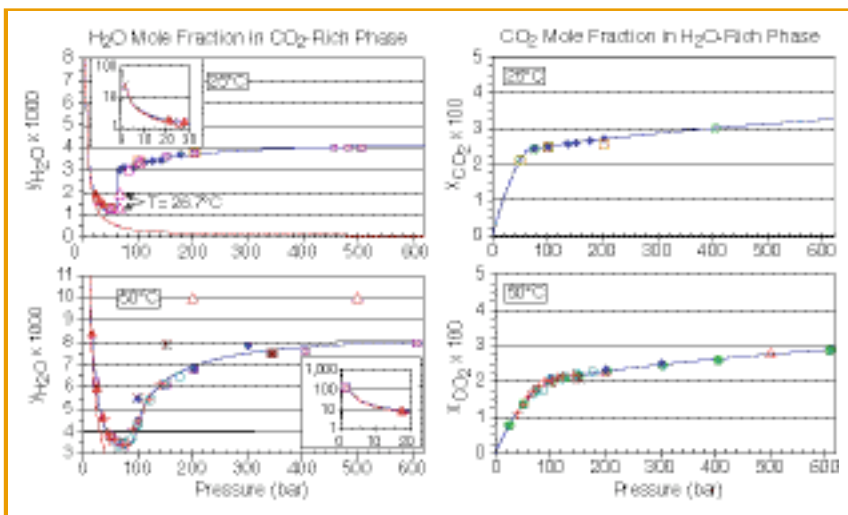

Figure 1. Calculated mutual solubilities of CO_2 and H_2O in this study (solid lines) and experimental data from the literature (symbols, with different shapes or colors for each different set of data). Dotted lines assume ideal mixing. Letters y and x refer to mole fractions in the compressed gas and aqueous phases, respectively.

requiring an iterative solution. Also, these models rely on a much smaller number of experimental data points. The noniterative procedure developed in this study reproduces experimental mutual solubilities of CO_2 and H_2O with sufficient accuracy for the study of geologic CO_2 disposal and enough simplicity to avoid degrading the performance of numerical fluid-flow simulations.

RELATED PUBLICATION

Spycher, N., K. Pruess, and J. Ennis-King, CO_2-H_2O mixtures in the geological sequestration of CO_2. I. Assessment and calculation of mutual solubilities from 12 to 100°C and up to 600 bar. Geochimica et Cosmochimica Acta, 2003 (in press); Berkeley Lab Report LBNL-50991, 2002.

ACKNOWLEDGMENTS

CARBON CYCLING IN THE SOUTHERN GREAT PLAINS: THE ARM/LBNL CARBON PROJECT

Margaret S. Torn, M.L. Fischer, William J. Riley, I. Pesenson, and J. Berry

Contact: Margaret S. Torn, 510/495-2223, mstorn@lbl.gov

RESEARCH OBJECTIVES

One of the challenges in carbon cycle research is the vast range of scales, from plants to continents, that must be bridged with measurements and models. The Atmospheric and Radiation Measurement (ARM)/LBNL Carbon Project is making a coordinated suite of carbon concentration, isotope, and flux measurements to support a range of scaling and integration exercises, including those proposed for the North American Carbon Program:

- Quantify the regional atmospheric CO_2 budget.
- Predict carbon fluxes, and the effect of land use and climate on them.
- Link local processes to regional and global models.

APPROACH

We are working at the DOE ARM Southern Great Plains testbed, a GCM-grid sized area centered in Northern Oklahoma. The carbon cycle data streams that our group produces are centered at the 60 m tower of the ARM Central Facility and include: precise CO_2 concentration profiles; carbon eddy covariance fluxes; National Oceanic and Atmospheric Administration–Climate Monitoring Diagnostics Laboratory (NOAA-CMDL) flasks in the mixed layer and free troposphere; and diurnal profiles of ^{13}C and ^{18}O in CO_2. In the next year, we are adding continuous CO measurements and flask sampling for $^{14}CO_2$ to assist with source attribution.

ACCOMPLISHMENTS

The continuous precise CO_2 measurements and the NOAA flask sampling tie the ARM testbed to the global atmospheric network. We observe that CO_2 concentrations are higher than global average for the latitude, reflecting continental sources. The diurnal cycle also shows the influence of continental atmospheric and terrestrial processes. At night (as seen in Figure 1), soil respiration and stable conditions lead to large buildups in CO_2, especially near the surface. During the day-

time, the atmosphere is well mixed and photosynthesis reduces atmospheric CO_2 concentrations. The highest concentrations of CO_2 are on nights of low wind speed and low mixing height. Our flux studies indicate that land use is the most important driver of spatial heterogeneity in fluxes in the study area. Using these results, regional-scale estimates of carbon fluxes based on "top-down" (atmospheric concentrations) and "bottom up" (distributed modeling and eddy flux measurements) approaches are in progress.

SIGNIFICANCE OF FINDINGS

Observations of Atmospheric CO_2 concentrations can improve estimates of surface fluxes, source types, and total atmospheric CO_2 stocks. However, the diurnal cycle of CO_2 concentration at continental sites reflects not only net ecosystem exchange, but also atmospheric mixing (planetary boundary layer) advection from surface winds, and anthropogenic carbon sources. Thus, interpreting atmospheric concentrations also requires understanding atmospheric processes and anthropogenic activity.

The isotopic and molecular composition of atmospheric samples creates fingerprints of fossil, biomass burning, and biogenic sources of carbon cycle gases. Our ongoing research efforts are focused on joining models of isotopic and atmospheric processes with land use patterns, and testing output against data covering multiple spatial and temporal scales.
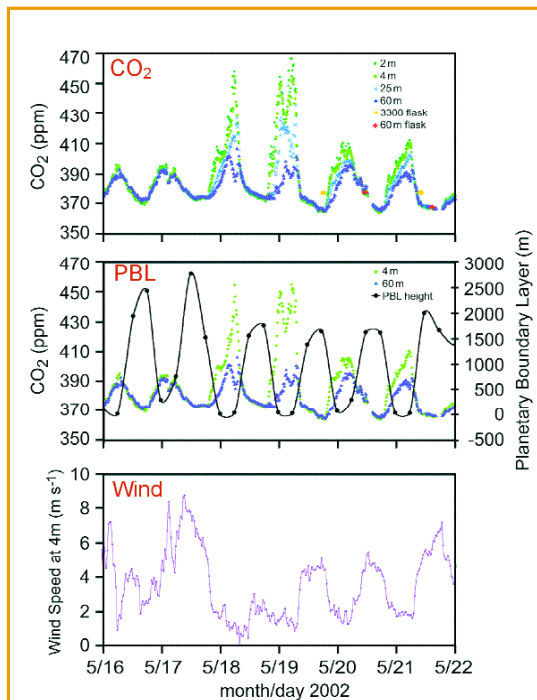

Figure 1. Diurnal cycle of CO_2, PBL height, and wind speed, May 16–22, 2002. Data were collected at the ARM Central Facility: CO_2 concentrations are from the continuous precise system at 60 m tower; red and yellow symbols are NOAA-CMDL flask data. PBL height was estimated from radiosonde profiles. Windspeed was measured at 4 m at the base of tower.

RELATED PUBLICATION

ARM Carbon Web Site: http://esd.lbl.gov/ARMCarbon/

ACKNOWLEDGMENTS

THE IMPORTANCE OF BELOWGROUND PLANT ALLOCATION FOR TERRESTRIAL CARBON SEQUESTRATION

Margaret S. Torn, T.E. Dawson[1], J.A. Bird, J. Gaudinski, and D.E. Williard
[1]University of California, Berkeley
Contact: Margaret S. Torn, 510/495-2223, mstorn@lbl.gov

RESEARCH OBJECTIVES

One strategy proposed for sequestering carbon in terrestrial ecosystems is to increase allocation of carbon to roots, on the assumption that root inputs are efficiently converted to stable soil organic matter (SOM). We are conducting research to fill critical gaps in understanding belowground carbon cycling and sequestration in soils of temperate forests, by characterizing:

- The lifetime of fine roots and implications for belowground net primary productivity
- Decomposition dynamics of root and needle/leaf C inputs
- Total residence time of belowground C, including SOM

APPROACH

The total residence time of root C in the ecosystem depends on how long roots live, the decay rate of root litter, the fraction of C inputs humified rather than lost as CO_2, and the stability of decomposition products. We are characterizing these aspects of residence time with field experiments and isotope-based approaches, including: (1) Natural abundance ^{14}C to determine fine-root lifetimes; (2) Litterbags and *in situ* incubations of ^{13}C-labeled litter to estimate litter decay rates; (3) ^{13}C-labeled litter to track root and needle decay into soil-respired CO_2, microbial biomass, and soil organic fractions.

We are conducting the full suite of measurements at a mature ponderosa pine forest (Blodgett, California), measuring roots and litter decay at Harvard Forest, Massachusetts, and focusing on fine-root lifetime at additional forests in the U.S. and Europe. We report here the first year's results on litter decomposition.

ACCOMPLISHMENTS

Several lines of evidence indicate that roots decompose more slowly than leaves or needles in the first year of decay. First, the litter bags had greater mass loss of needles compared to roots at Blodgett (75% versus 90% mass remaining, respectively) and leaves compared to roots at Harvard Forest (65% versus 90% mass remaining, respectively). Second, for the ^{13}C-labeled litter, recovery of fine-root C exceeded that of needles (77% versus 52%, respectively). Third, after one year, more of the needle material had been reduced to a smaller size class (<2 mm) than had the roots (42% and 24%, respectively), see figure 1. Finally, the loss of C as respired CO_2 was greater for needles than for roots.

Decay rates may vary because of chemical quality or the depth of placement, with the latter affecting both microclimate and soil mineral composition. We saw little or no significant effect on decay rates

between two depths (O and A horizons), for either the litter bag or the ^{13}C label experiments. In contrast, we did find a greater proportion of C compounds associated with recalcitrance, such as acid-soluble C and lignin, in roots compared to needles and leaves. Preliminary results suggest that chemical quality, rather than timing or depth of input to soil, was responsible for the differences in decay rates.
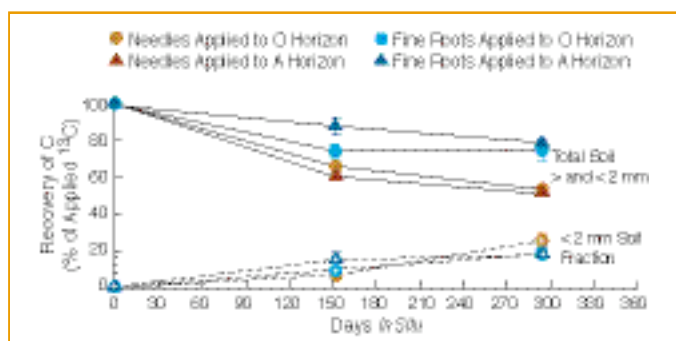

Figure 1. Percent C recovery from needle and root litter after 10 months *in situ*. Litter was applied to the top of the O horizon or 2–5 cm below the O/A interface in the A horizon during November 2001. Shown are total C recovery in the whole soil (solid lines) and the <2 mm fraction (dashed lines). Means (n = 4) and standard errors are shown.

SIGNIFICANCE OF FINDINGS

Although roots live longer and decay more slowly than leaves or needles in these temperate forests, estimating the long-term sequestration potential will require a second phase of research on the humification pathways and stabilization of root inputs.

RELATED PUBLICATION

Torn, M.S., S. Davis, J.A. Bird, M.R. Shaw, and M.E. Conrad, Automated analysis of $^{13}C/^{12}C$ ratios in CO_2 and dissolved inorganic carbon for ecological and environmental applications. Rapid Communications in Mass Spectrometry, 2003 (in press). Berkeley Lab Report LBNL-53147.

ACKNOWLEDGMENTS

A Whole-Forest Pulse-Label Study of Microbial Dynamics and Root Turnover

Margaret S. Torn, J. Gaudinski, K. Treseder[1], J. Westbrook, D. Joslin[2], and C. Swanston[3]
[1]University of Pennsylvania, Philadelphia, PA
[2]Belowground Forest Research, Oak Ridge, TN
[3]Lawrence Livermore National Laboratory, Livermore, CA
Contact: Margaret S. Torn, 510/495-2223, mstorn@lbl.gov

RESEARCH OBJECTIVES

In the summer of 1999, there was a large atmospheric release of $^{14}CO_2$ near the Oak Ridge Reservation (ORR), Tennessee, from a local incinerator. The rapid photosynthetic uptake of the $^{14}CO_2$ created a pulse label for studying carbon (C) cycling through the ORR forests. Several institutions (four DOE labs, two UC campuses, the U.S. Forest Service, and a private firm) are utilizing this whole-ecosystem isotopic label to address unresolved issues in terrestrial carbon cycling. At Berkeley Lab, we are investigating (1) the longevity of fine roots, and (2) the pathways from leaf and root inputs to microbial biomass and soil organic matter. Here, we summarize the first year's results.

APPROACH

The ORR team used enriched and near-background leaf litter to create four treatments, depending on plant ^{14}C inputs: enriched roots, enriched leaf litter, both enriched leaves and roots, and no enrichment. We determined fine-root longevity by tracing the radiocarbon label through live and dead root populations. New root growth was isolated by harvesting roots that grew through a screen in the soil. For fungal dynamics, ectomycorrhizae were hand-picked from freshly harvested roots. For microbial biomass, chloroform fumigation-extracts of soils were freeze-dried and combusted for graphitization. Radiocarbon content was measured at Lawrence Livermore National Laboratory by accelerator mass spectrometry.

ACCOMPLISHMENTS AND SIGNIFICANCE

New root growth contains about 10–20% of the ^{14}C signature from the previous year's growth (Figure 1), showing that roots grow from a mixture of recent photosynthate and C storage from the previous year. This result is helping us parameterize root models and estimate fine-root turnover based on atmospheric trends in $^{14}CO_2$. Our data from this and other forests show no correlation in root lifetimes between different root-diameter size classes; we believe that the diameter-lifetime correlation observed by minirhizotron studies may only be valid for roots living less than 1 year.

The only plots in which ectomycorrhizal fungi were ^{14}C-enriched were those with trees that had enriched roots. This shows that the fungi were not receiving carbon from decom-

position of organic matter, but rather from the live roots they colonized.

Microbial biomass was enriched in all treatments and depths. The rapid enrichment (~500‰ in 2002) is consistent with conceptual models of microbes as an active carbon pool that decomposes roots, litter, and dissolved organic material. The similarity to heterotrophic respiration values (~400–600‰) suggests that we may be able to use microbial biomass ^{14}C to estimate the season-integrated signature soil-respired $^{14}CO_2$. We are using results from these studies to improve models of forest C cycling and sequestration.

RELATED PUBLICATION

Enriched Background Isotope Study (EBIS) Workshop Report, January 2003; www.esd.ornl.gov/programs/WBW/EBISwkshp2003.htm.
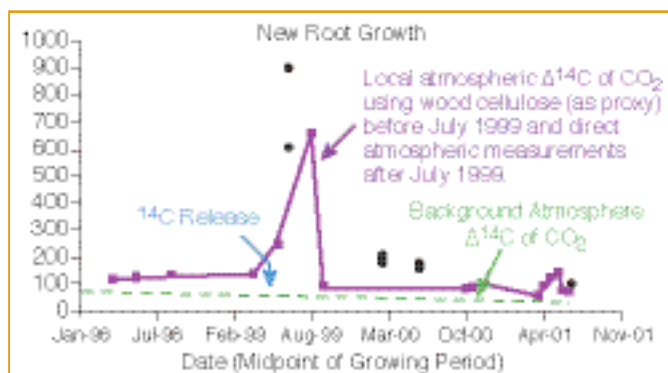
ACKNOWLEDGMENTS

Figure 1. The change in $\Delta^{14}C$ of new root growth (black circles) over time at Walker Branch, Oak Ridge, TN. The purple line shows atmospheric $^{14}CO_2$ at Oak Ridge, with the major ^{14}C release in 1999, based on wood cellulose before July 1999 and direct atmospheric measurements after July 1999. The dashed green line is background atmospheric $\Delta^{14}CO_2$.

TOUGHREACT Simulation Studies for Mineral Trapping Following CO₂ Disposal in Deep Saline Aquifers

Tianfu Xu, John A. Apps, and Karsten Pruess

Contact: Tianfu Xu, 510/486-7057, tianfu_xu@lbl.gov

RESEARCH OBJECTIVES

A reactive fluid flow and geochemical transport numerical model TOUGHREACT has been developed for evaluating long-term CO_2 disposal in deep geologic formations. The model is needed because alteration of the aluminosilicate minerals in the predominant host rock is very slow and not amenable to laboratory experiment under ambient deep-formation conditions. Using this model, we performed: (1) batch geochemical modeling analysis with three rock types (glauconitic sandstone from the Canada Alberta Sedimentary Basin, a sediment from the United States Gulf Coast, and a dunite); (2) reactive transport simulations of a 1-D radial-well region under CO_2 injection conditions to analyze CO_2 immobilization through carbonate precipitation in Gulf Coast sandstone of the Frio formation (Texas); and (3) reactive transport simulations of CO_2 sequestration in bedded sandstone-shale sequences.

APPROACH

TOUGHREACT was developed by introducing reactive chemistry into the framework of the existing multiphase fluid and heat flow code TOUGH2. The code can be applied to one, two-, or three-dimensional porous and fractured media with physical and chemical heterogeneity, and can accommodate any number of chemical species present in liquid, gas, or solid phases. A wide range of subsurface thermal-physical-chemical processes is considered, including (1) redox processes that could be important in deep subsurface environments; (2) the presence of organic matter; (3) the kinetics of chemical interactions between the host rock minerals and the aqueous phase; and (4) CO_2 solubility dependence on pressure, temperature, and salinity of the system.

ACCOMPLISHMENTS

We evaluated the geochemical evolution under both natural background and CO_2 injection conditions. Changes in porosity were monitored during the simulations. Under favorable conditions, the amount of CO_2 that may be sequestered by precipitation of secondary carbonates is comparable to (and can be larger than) the effect of CO_2 dissolution in pore waters. The addition of CO_2 mass as secondary carbonates (Figure 1) to the solid matrix decreases porosity, and a small porosity decrease can result in a significant decrease in permeability. The mass-transfer pattern of aqueous chemical components

under high CO_2 pressure conditions is different from that under natural conditions—most CO_2 sequestration occurs in the sandstone. Simulation results of mineral alteration under natural conditions agree well with field observations. The limited information currently available for the mineralogy of high-pressure CO_2 reservoirs is also generally consistent with our simulation. More details are given in Xu et al. (2003a, b, c).

RELATED PUBLICATIONS

Xu, T, J. A. Apps, and K. Pruess, Numerical simulation to study mineral trapping for CO_2 disposal in deep aquifers. Applied Geochemistry, 2003 (in press).

Xu, T, J.A. Apps, and K. Pruess, Reactive geochemical transport simulation to study mineral trapping for CO_2 disposal in deep arenaceous formations. Journal of Geophysical Research, 108 (B2), 2071, doi:10.1029/2002JB001979, 2003.

Xu, T, J.A. Apps, and K. Pruess, Mass transfer, mineral alteration, and CO_2 sequestration in a sandstone-shale system. Chemical Geology, 2003 (submitted).
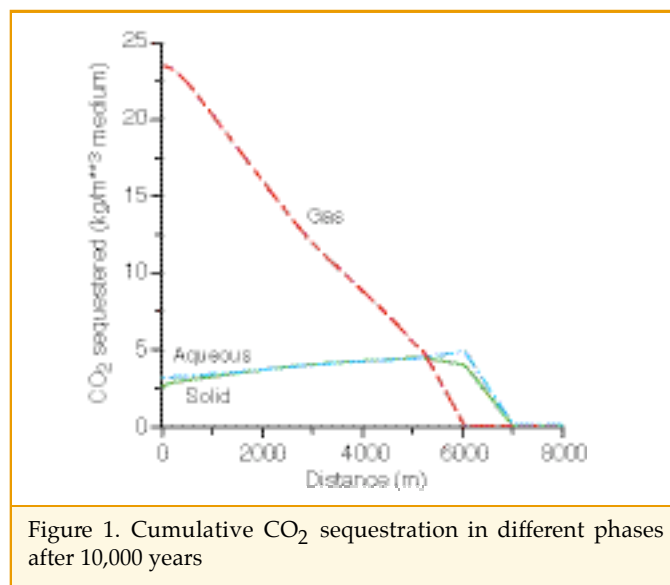
ACKNOWLEDGMENTS

Figure 1. Cumulative CO_2 sequestration in different phases after 10,000 years